Nonequilibrium Molecular Dynamics Simulation of the Thermocapillary Effect

Molekulardynamische Nichtgleichgewichtssimulation des thermokapillaren Effekts

Figures

Vom Fachbereich Maschinenbau an der Technischen Universität Darmstadt

zur Erlangung des akademischen Grades eines Doktor-Ingenieurs (Dr.-Ing.)

genehmigte DISSERTATION

vorgelegt von

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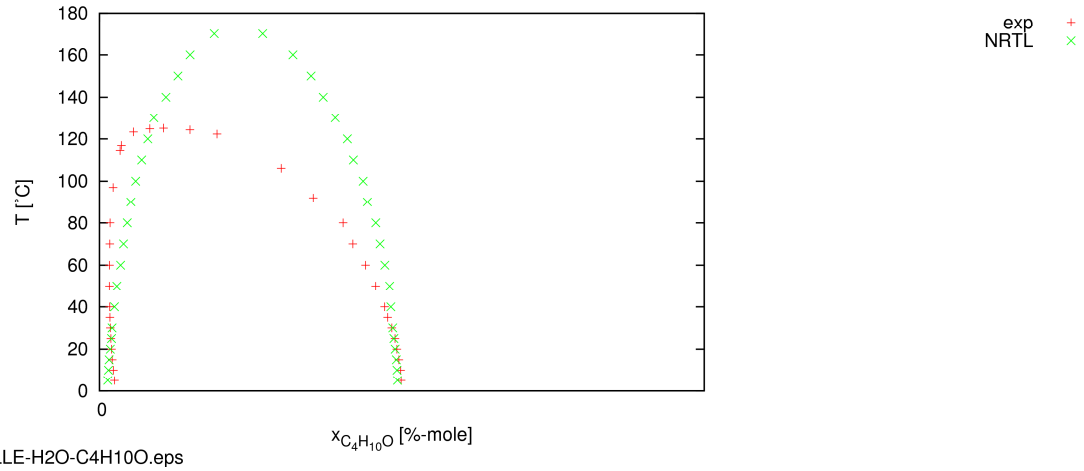
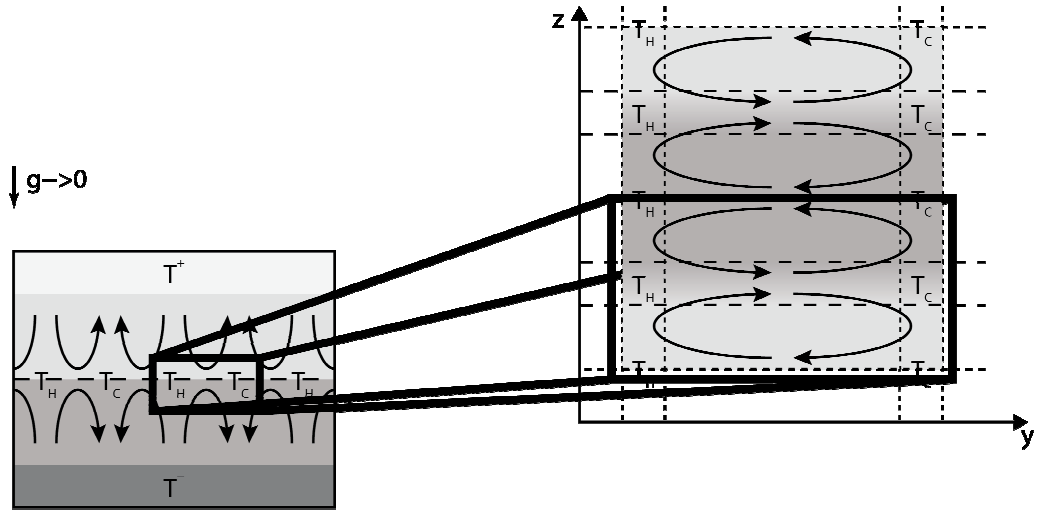
15 Figures



LLE-H2O-C4H10O.eps

Figure 1 **Liquid-liquid phase diagram of a binary water / 1-Butanol mixture at ambient pressure We show the data from experiments and from thermodynamic calculations using the NRTL equation [sorensen1979].**



a) Physical experiment: Thermocapillary convection in a binary system of two partially miscible liquids (green and yellow) at different bulk phase temperatures T_+ and T_- [oertel2002]; Temperature fluctuations in the interfacial region ignite the thermocapillary convection that selfsustains by dragging new hot liquid from the bulk phases to the interface where the latter gives off the heat to the counterflow in the opposite phase and returns into the hot bulk phase. Under certain system conditions the thermocapillary convection becomes stationary and temperature differences along the interface can be observed between the places where the convection approaches (T_H) and leaves (T_C). Note, gravity is necessary to keep the interface flat but buoyancy driven convection shall be negligible.

b) Numerical experiment: reproduction of the physical experiment in MD simulation with simplifications mentioned in the text; The hatching represents the interfacial region. In order to comply with the periodic system boundaries the thermocapillary system is mirrored along the *xy*- and the *xz*-plane.

Figure 2 **Thermocapillary convection in a physical experiment and its reproduction in a numerical experiment**

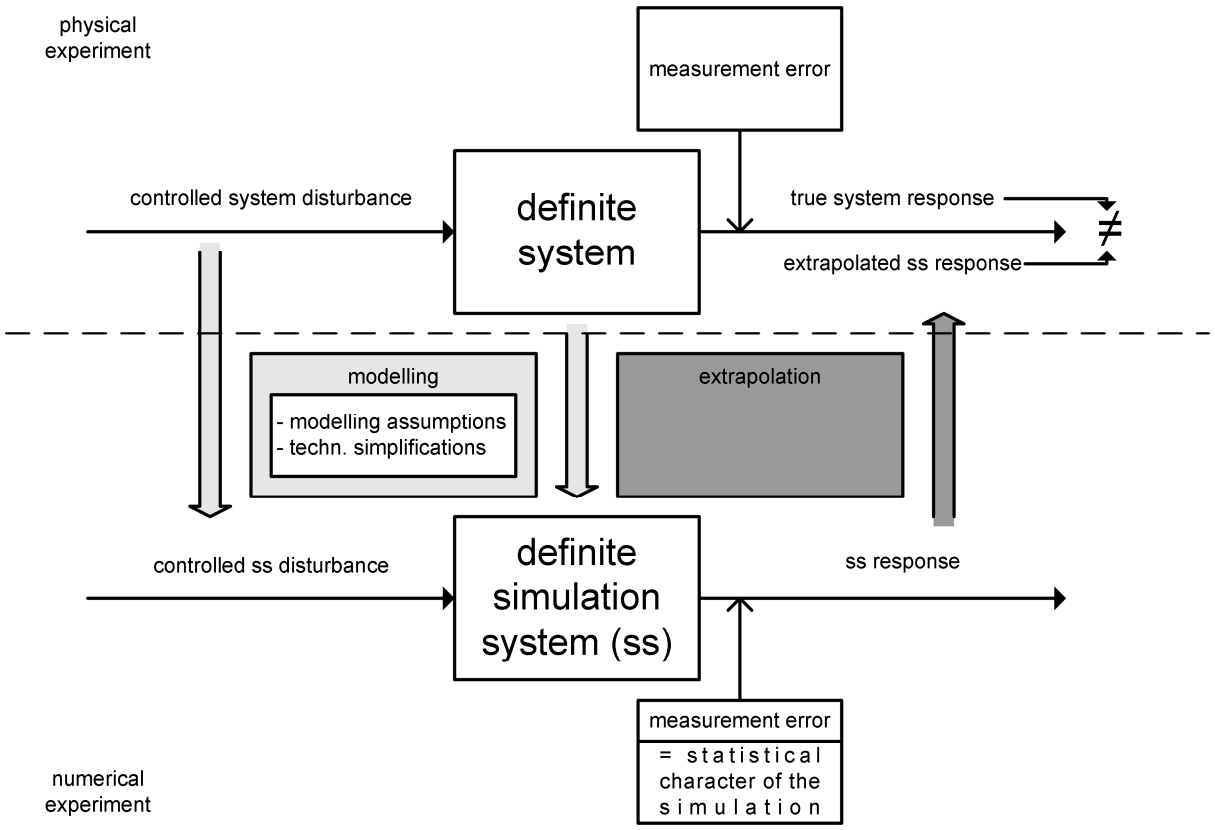


Figure 3 **Scheme of our procedure to study the thermocapillary effect in a "numerical experiment" instead of the "physical experiment"**

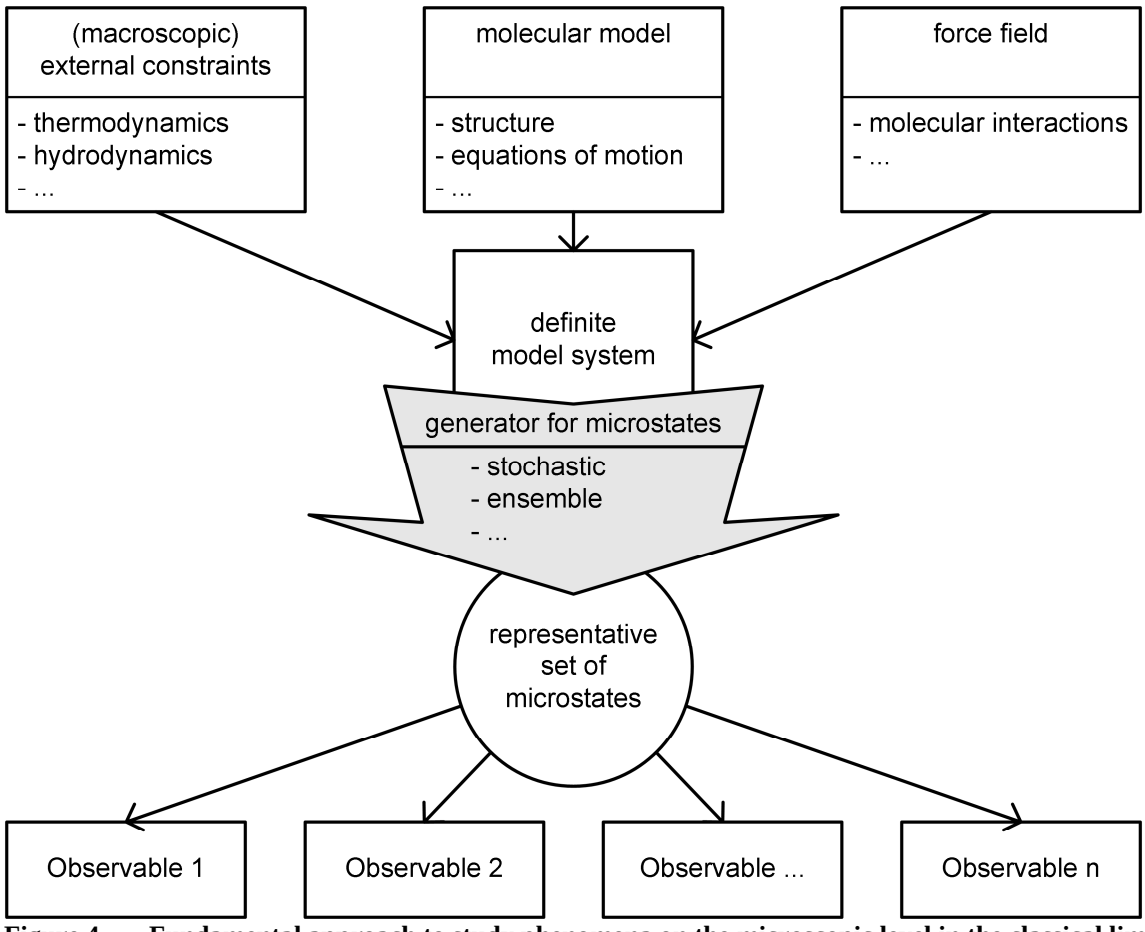


Figure 4 **Fundamental approach to study phenomena on the microscopic level in the classical limit**

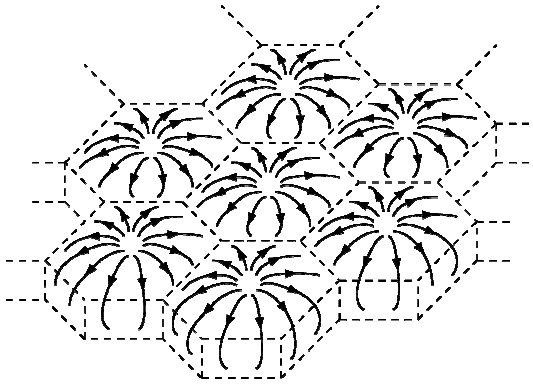


Figure 5 **Hexagonal convection pattern of the Marangoni-Bénard instability**

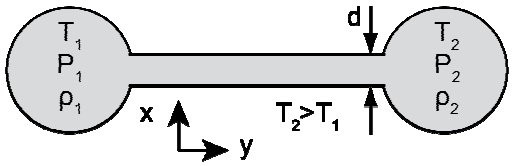


Figure 6 **Typical experimental setup for the study of thermal creep and molecular gas flows**

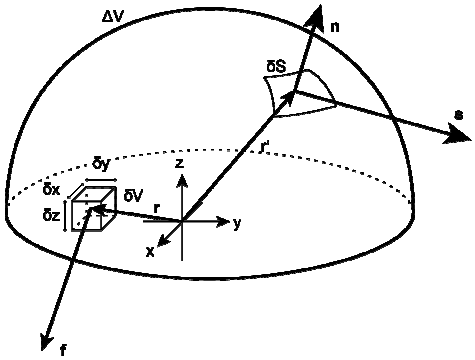


Figure 7 **Local surface and volume forces on a macroscopically small subvolume ΔV**

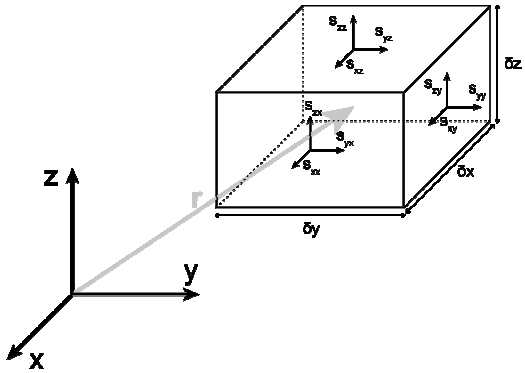
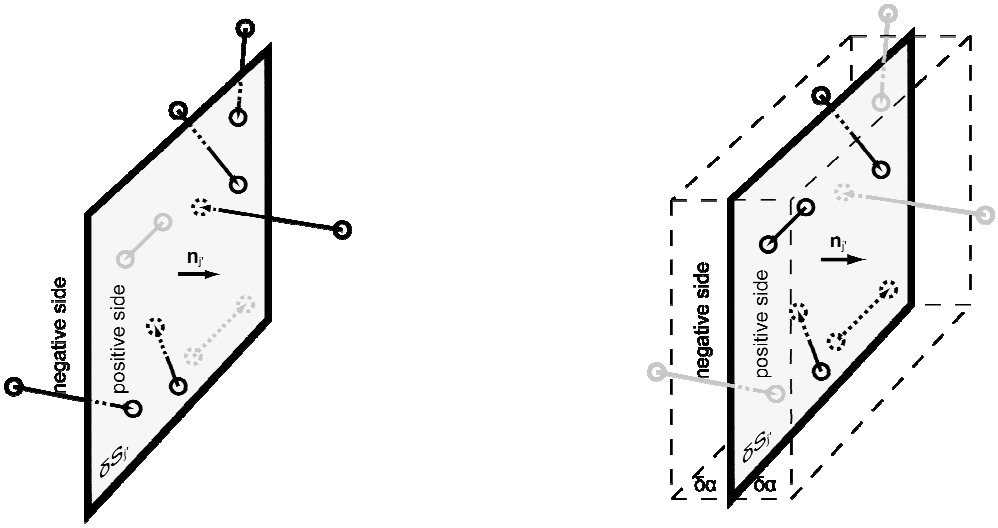


Figure 8 **Stress tensor elements for three mutually perpendicular and macroscopically small cut surfaces $\delta S_{j'}$**



a) definition, that considers only the particles crossing the cut surface

b) definition, that considers all particles in the vicinity of the cut surface

Figure 9 **Two different microscopic definitions of the kinetic contribution to the stress vector associated with the cut surface $\delta S_{j'}$**

Both sketches show the same exemplary particle arrangements at two subsequent time steps. Different particles contribute to the stress vector depending on the definition. We highlight them in black compared to the grey ones.

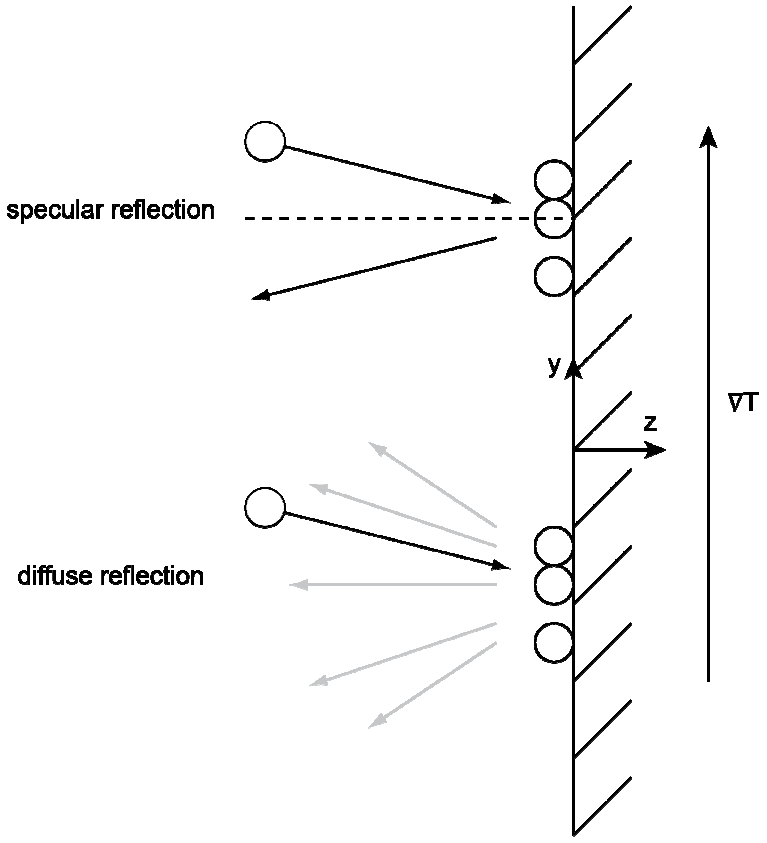


Figure 10 **Diffuse and specular reflection at a gas adsorption layer**

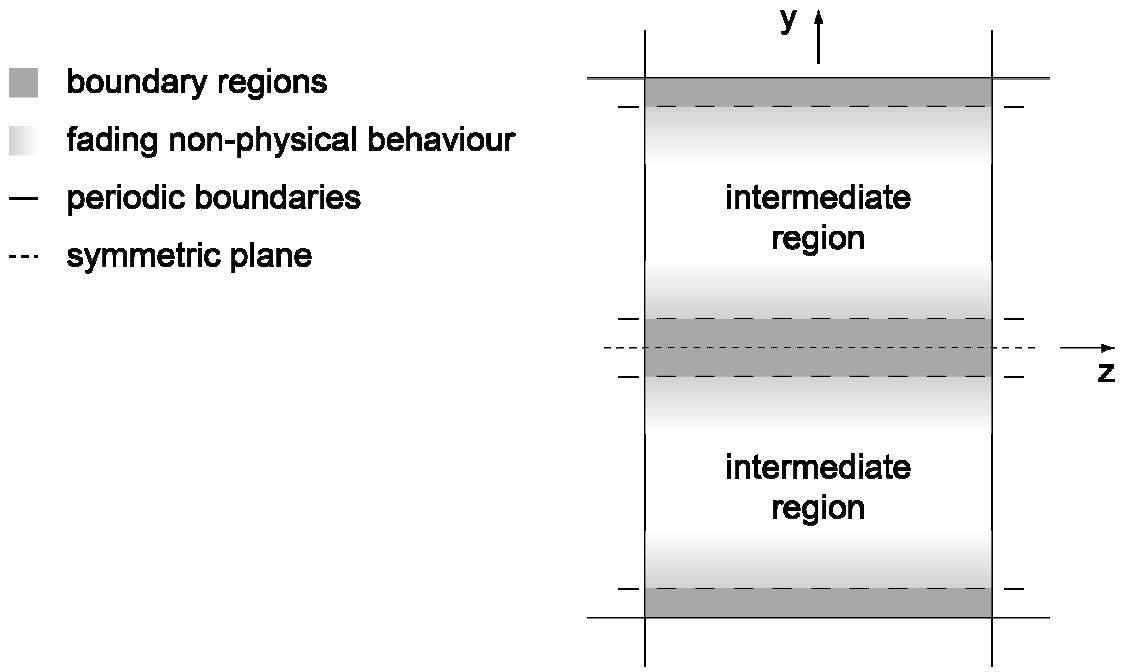


Figure 11 **Sketch of a typical BD NEMD simulation system with indication of the perturbed "boundary regions"**
In the "intermediate regions" between them, the non-physical behaviour in the perturbation fades

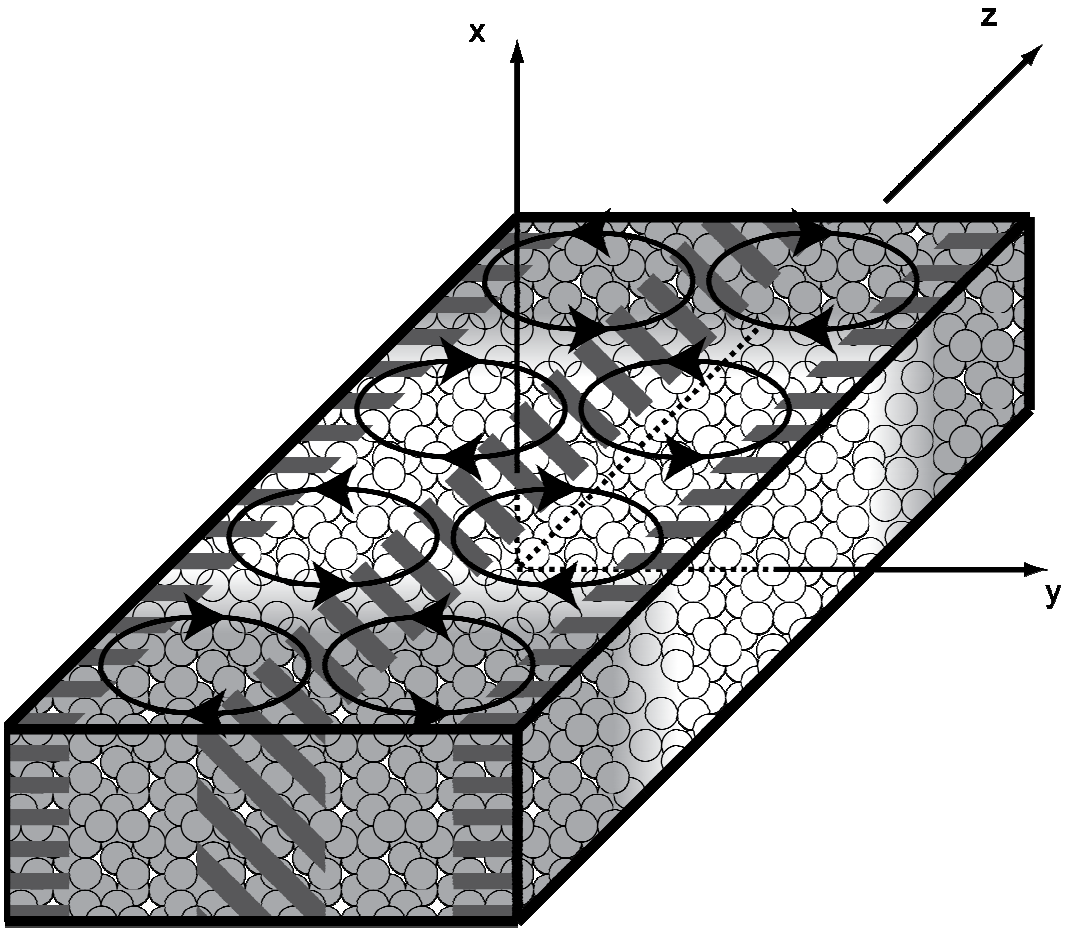


Figure 12 Setup of the simulation system with the expected convection roll cells driven by the thermocapillary effect

The grey and white circles represent the particles of the two species, generally called ArA and ArB here. The liquid-liquid interfaces are oriented parallel to the *xy*-plane. The regions parallel to the *yz*-plane represent the "thermostated regions". The hot one carries a horizontal and the cold one a diagonal hatching.

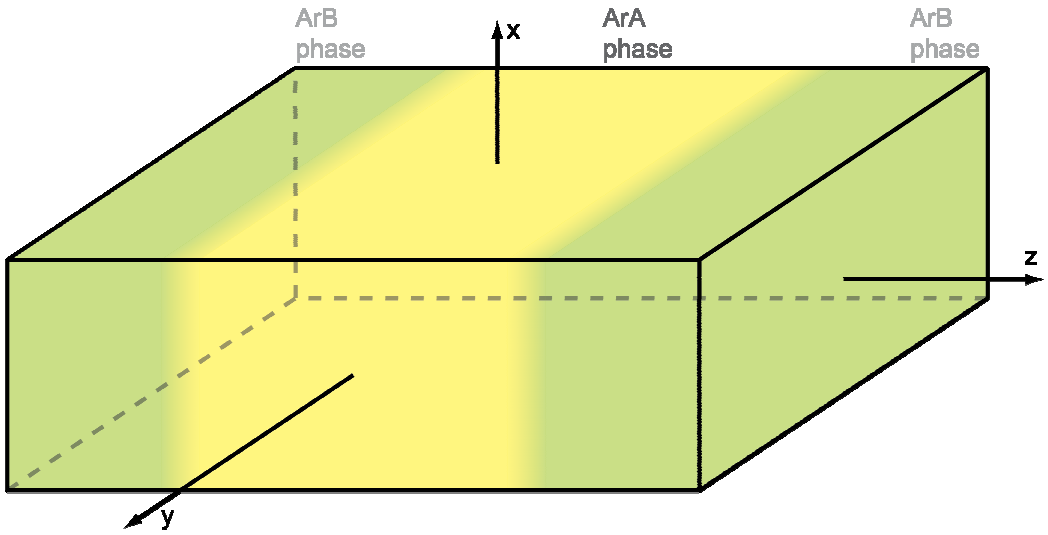


Figure 13 **Basic sketch of the equilibrium interfacial systems simulated**

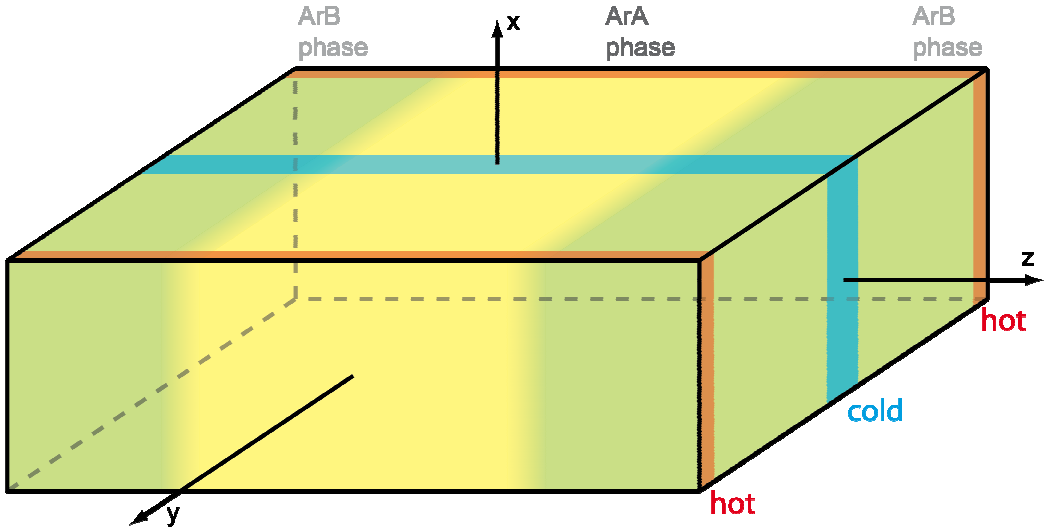
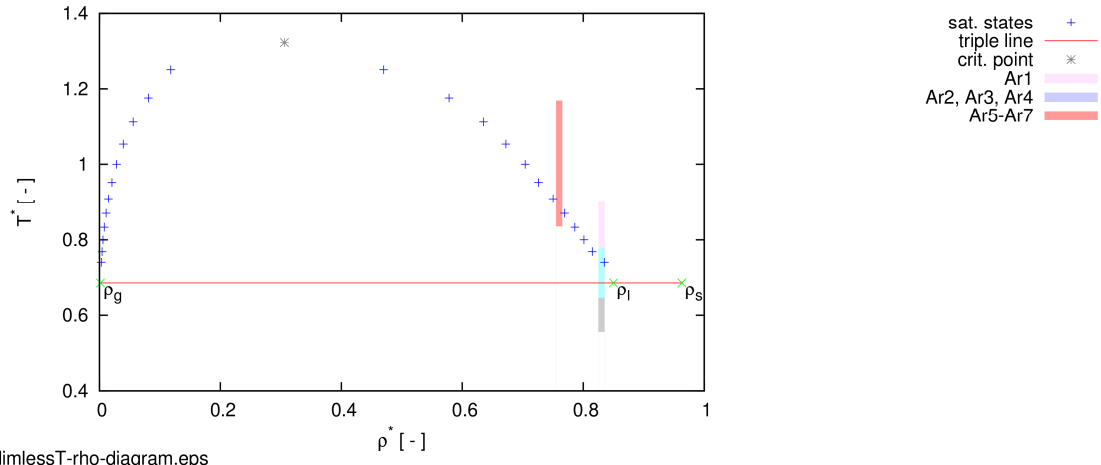


Figure 14 **Basic sketch of the nonequilibrium interfacial systems simulated**
It indicates additionally the boundary or thermostated regions.



dimlessT-rho-diagram.eps

Figure 15 **Dimensionless $T^*\rho^*$-diagram of the saturation curve of pure one-centre LJ particle substances**
The diagram shows additionally the reduced temperature-density ranges in the mixtures simulated in this work.

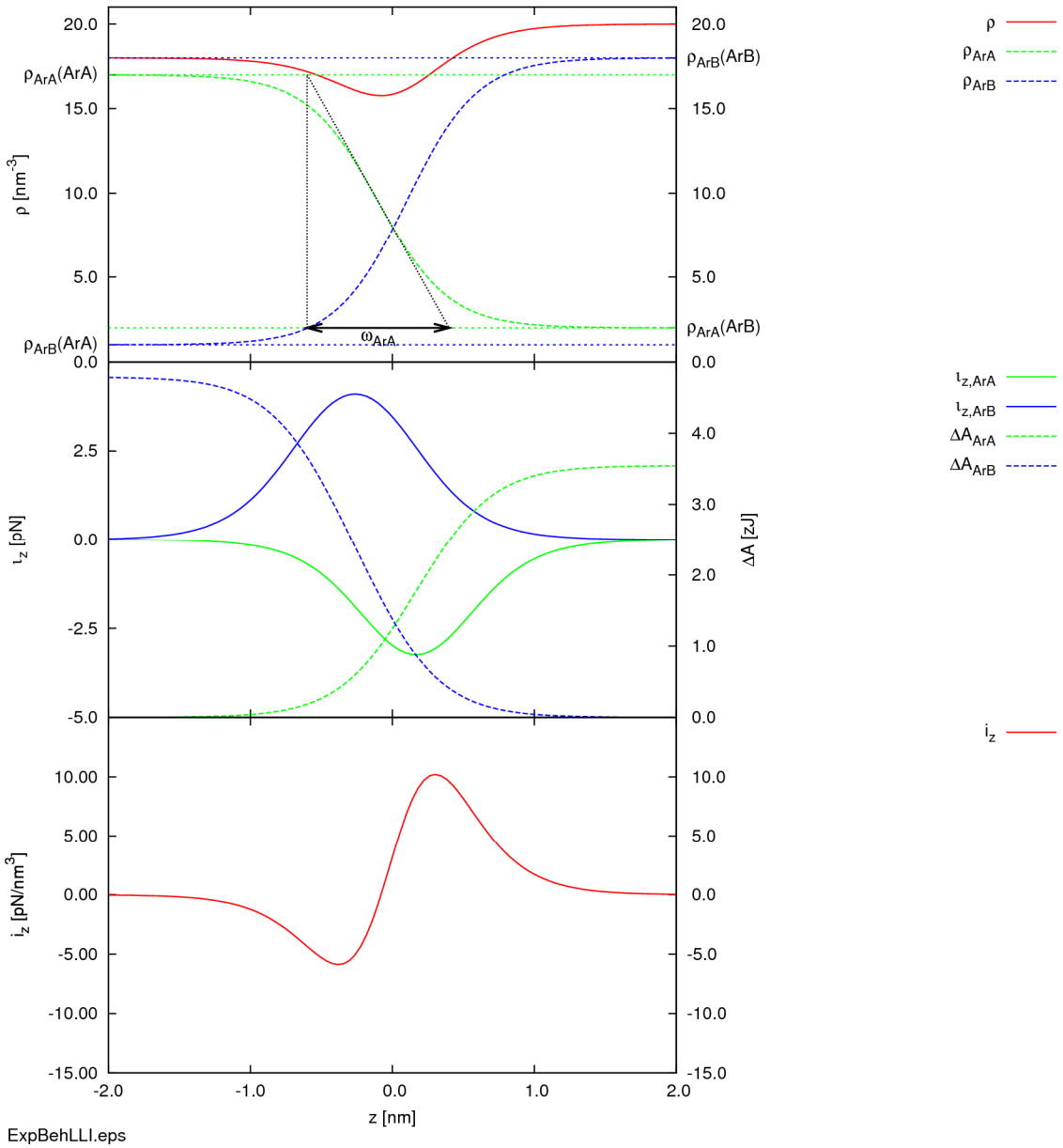
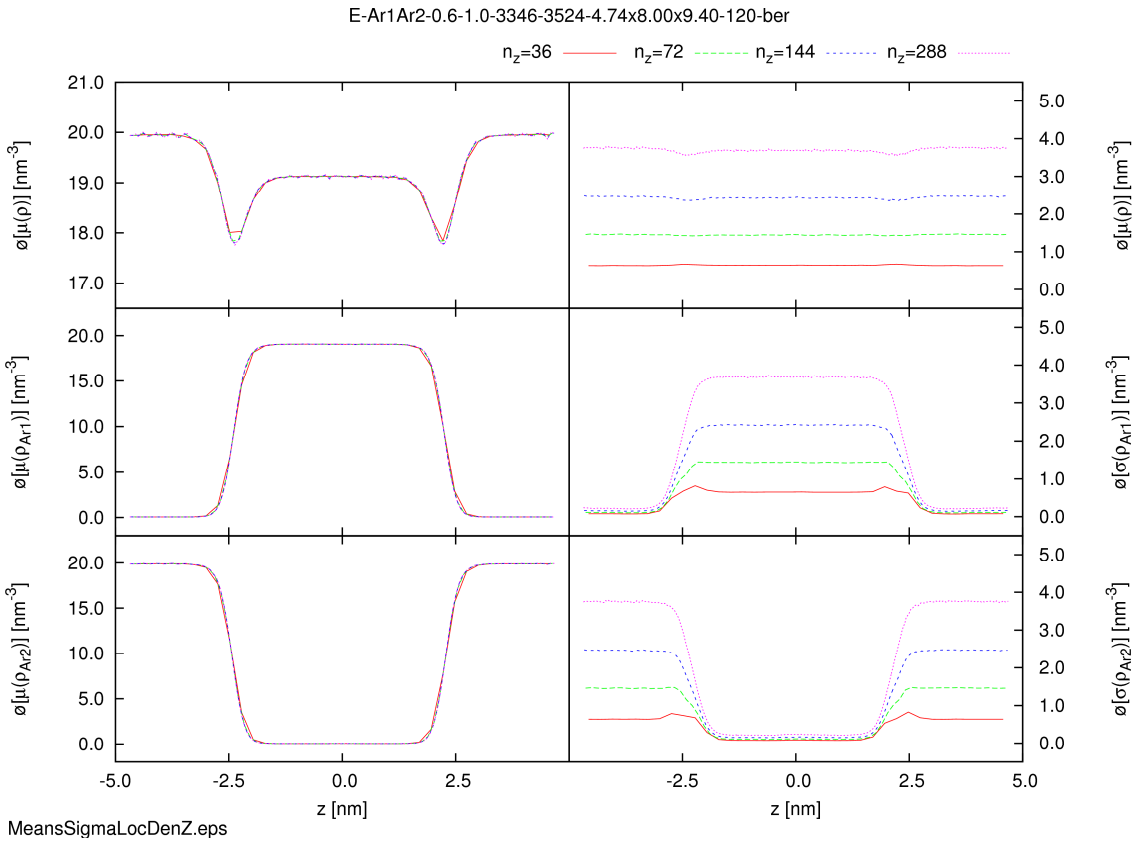


Figure 16 **Typical hyperbolic tangent partial density profiles (top) at a liquid-liquid interface in a mixture of the substances ArA and ArB**
We show additionally the related interatomic particle force and potential of mean force profiles (middle) as well as the related interatomic volume forces (bottom). The ArA rich phase resides on the left side and the ArB rich phase on the right side. The interfacial width related to the ArA partial density profile ω_{ArA} is indicated by the arrow.



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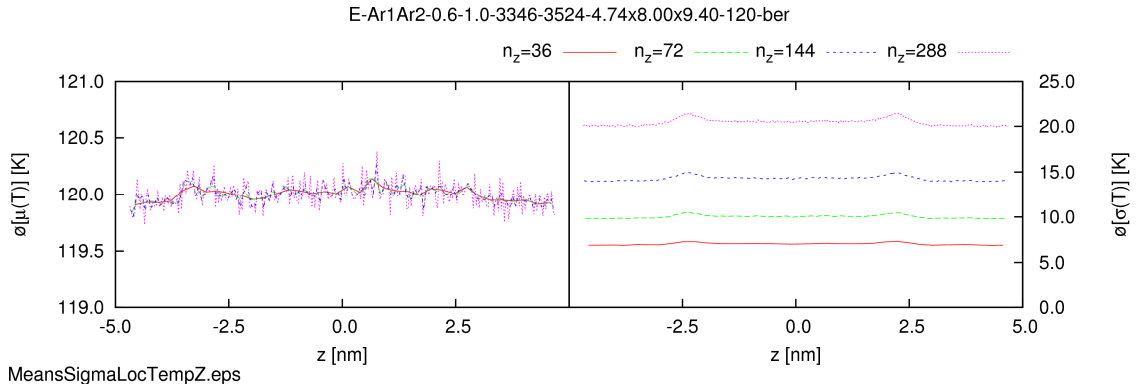
a) Densities



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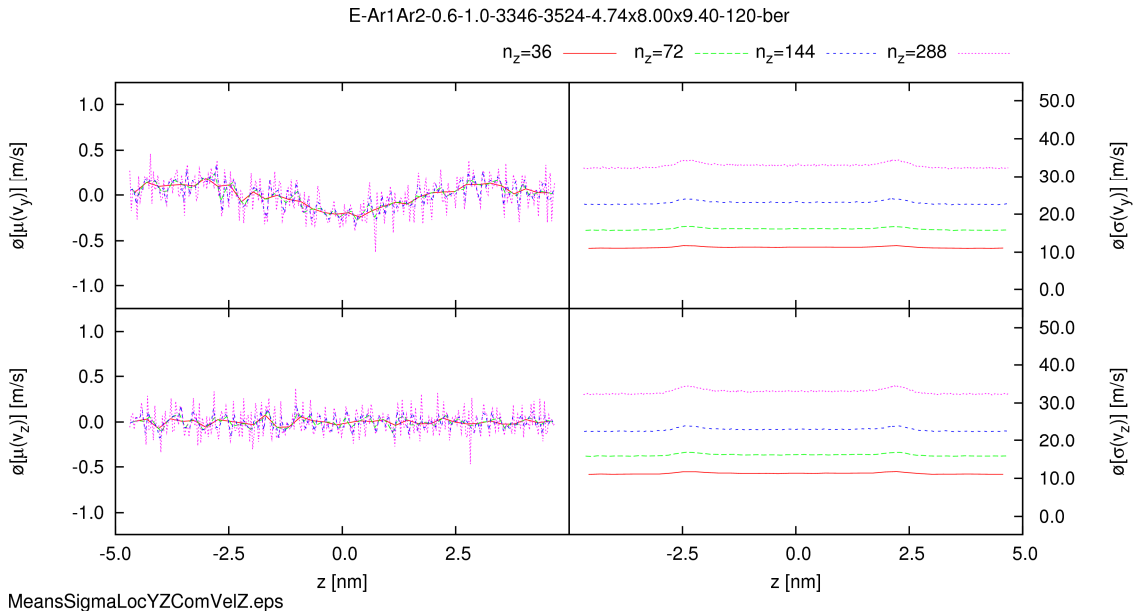
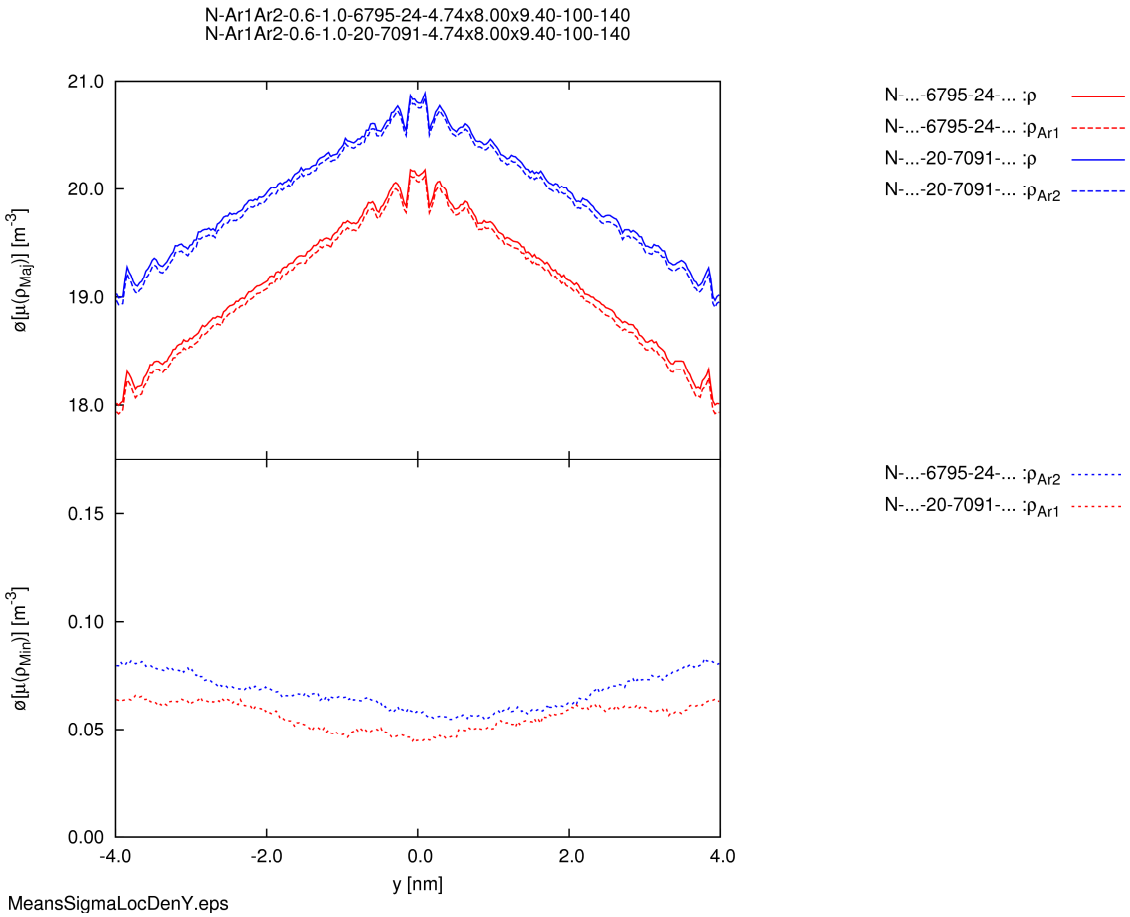
b) Temperatures



c) Com Velocities

Figure 17 Local observables obtained for different spatial resolutions in separate *NVT* simulations of the first heterophasic equilibrium interfacial system
We divide it into different numbers of slab-shaped subvolumes in the analysis. Nondefined results occur for a subdivision into 576 subvolumes since some of them are not populated by particles at every time step.



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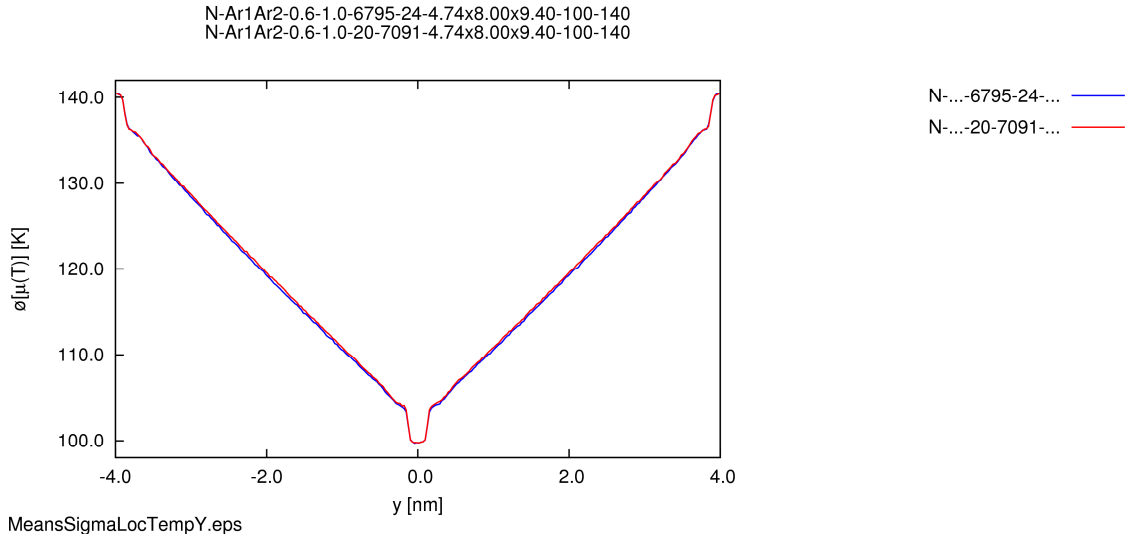
a) Densities



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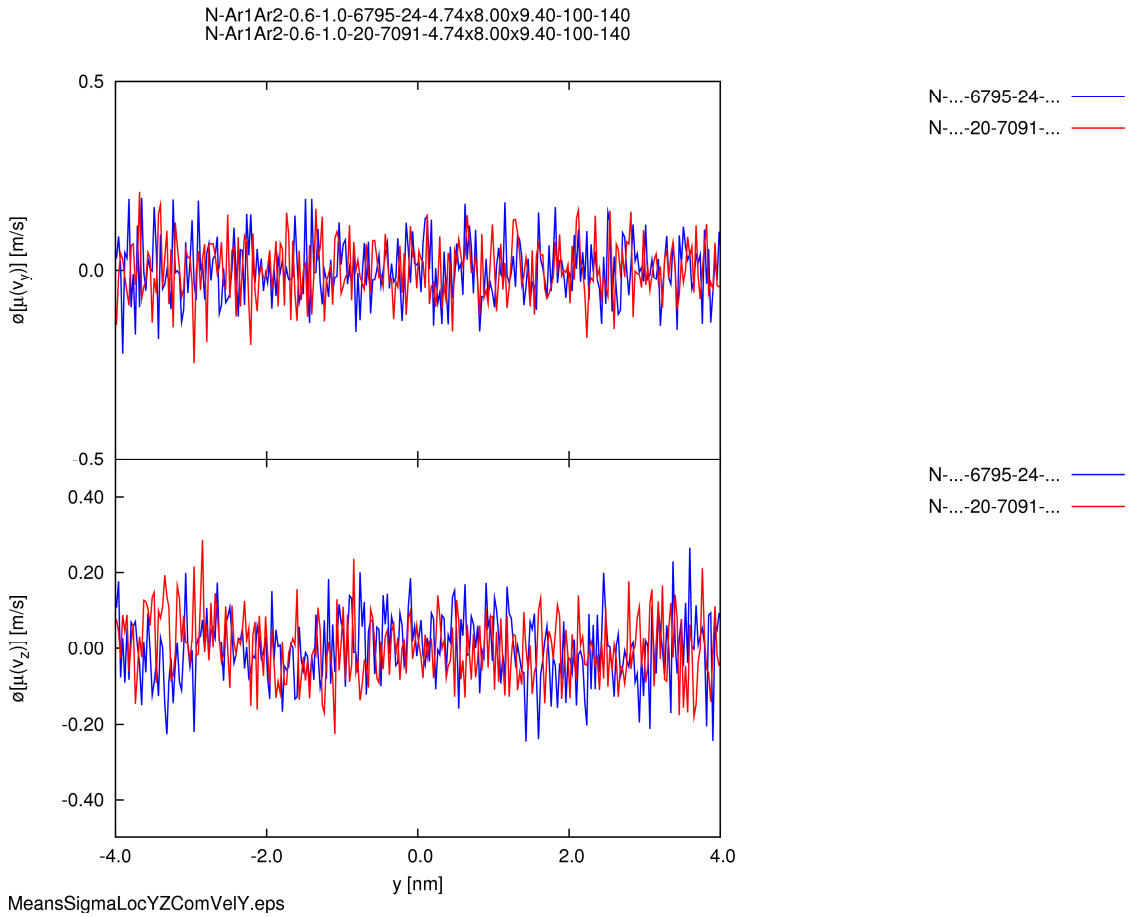
b) Temperatures



c) Com velocities

Figure 18 Comparison of the local observables obtained by using slabs in the two first nonequilibrium one-phase systems N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140

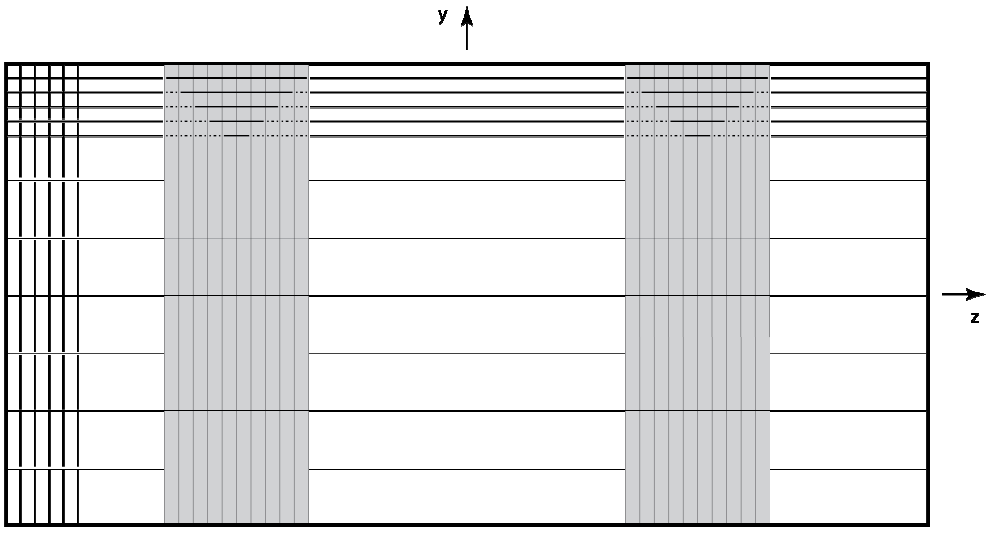


Figure 19 Locations and orientations of selected confined cut-surfaces used in the determination of the local stresses

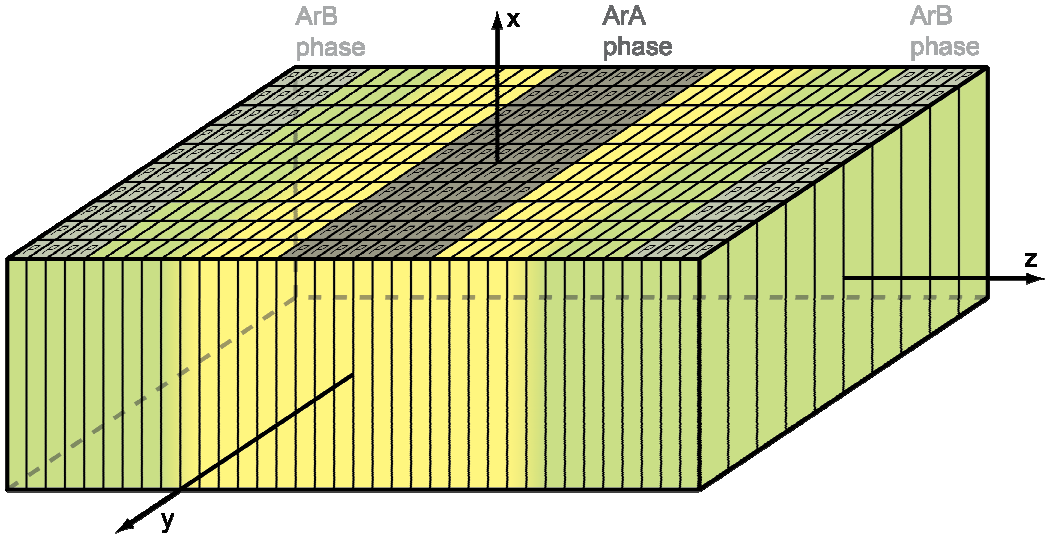


Figure 20 **Sketch of an equilibrium interfacial system together with the subvolumes for which the local observables are computed**
The subvolumes lumped together into the ArA or ArB phases are indicated in different shades of grey. The subvolumes considered in the calculation of the P-averages in each phase are additionally labelled with the letter "P".

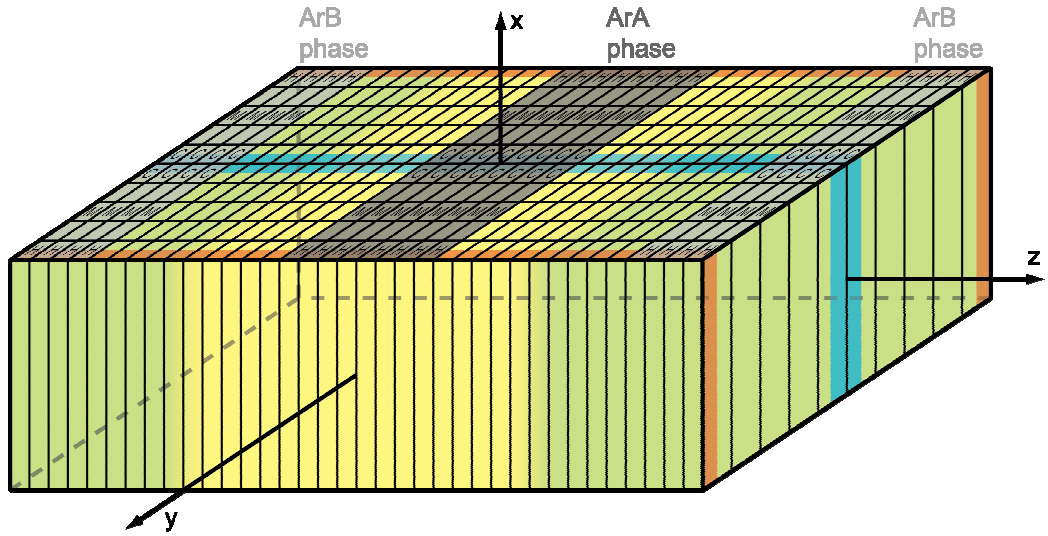
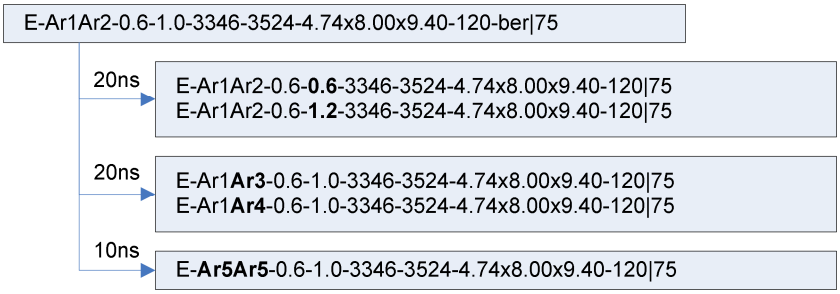
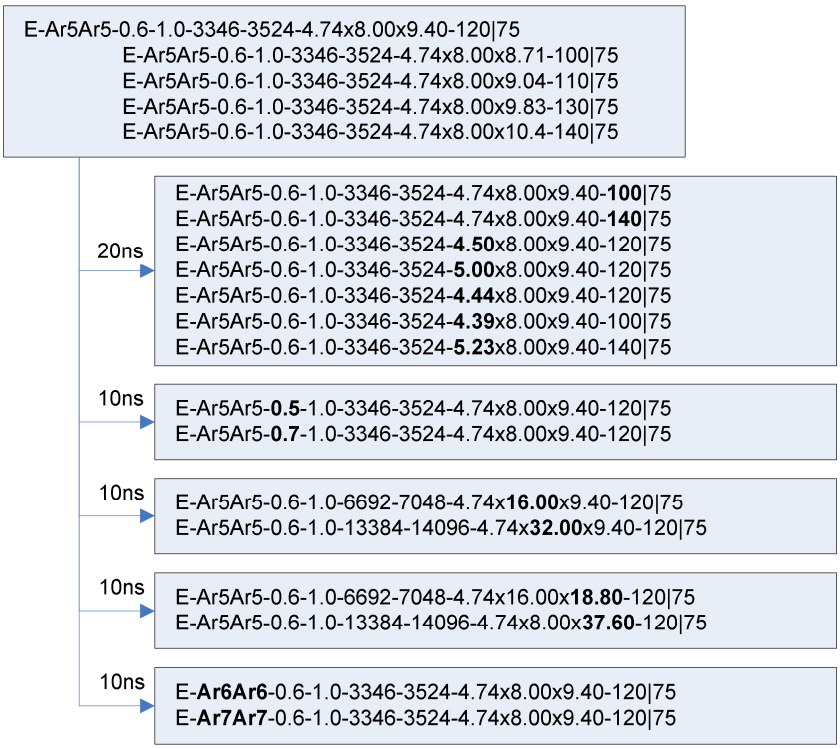


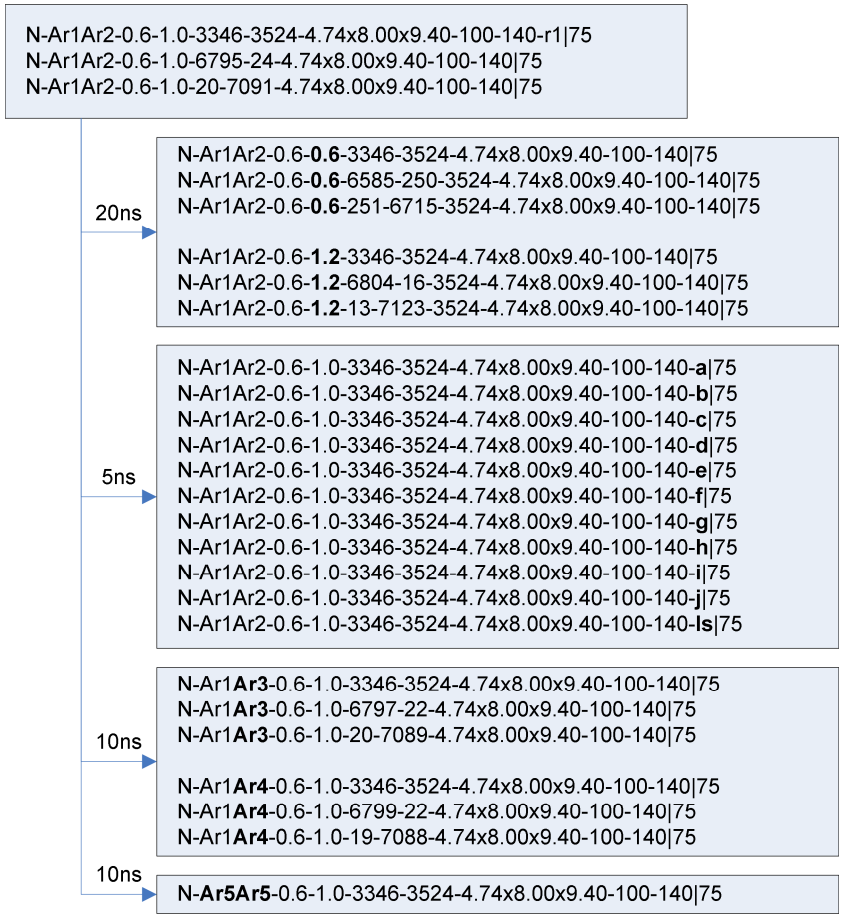
Figure 21 **Sketch of a nonequilibrium interfacial system together with the subvolumes for which the local observables are computed**
The subvolumes lumped together into the ArA or ArB phases are indicated in different shades of grey. The subvolumes considered in the calculation of the C, M, or H-averages in each phase are labelled with the letters "C", "M", or "H".



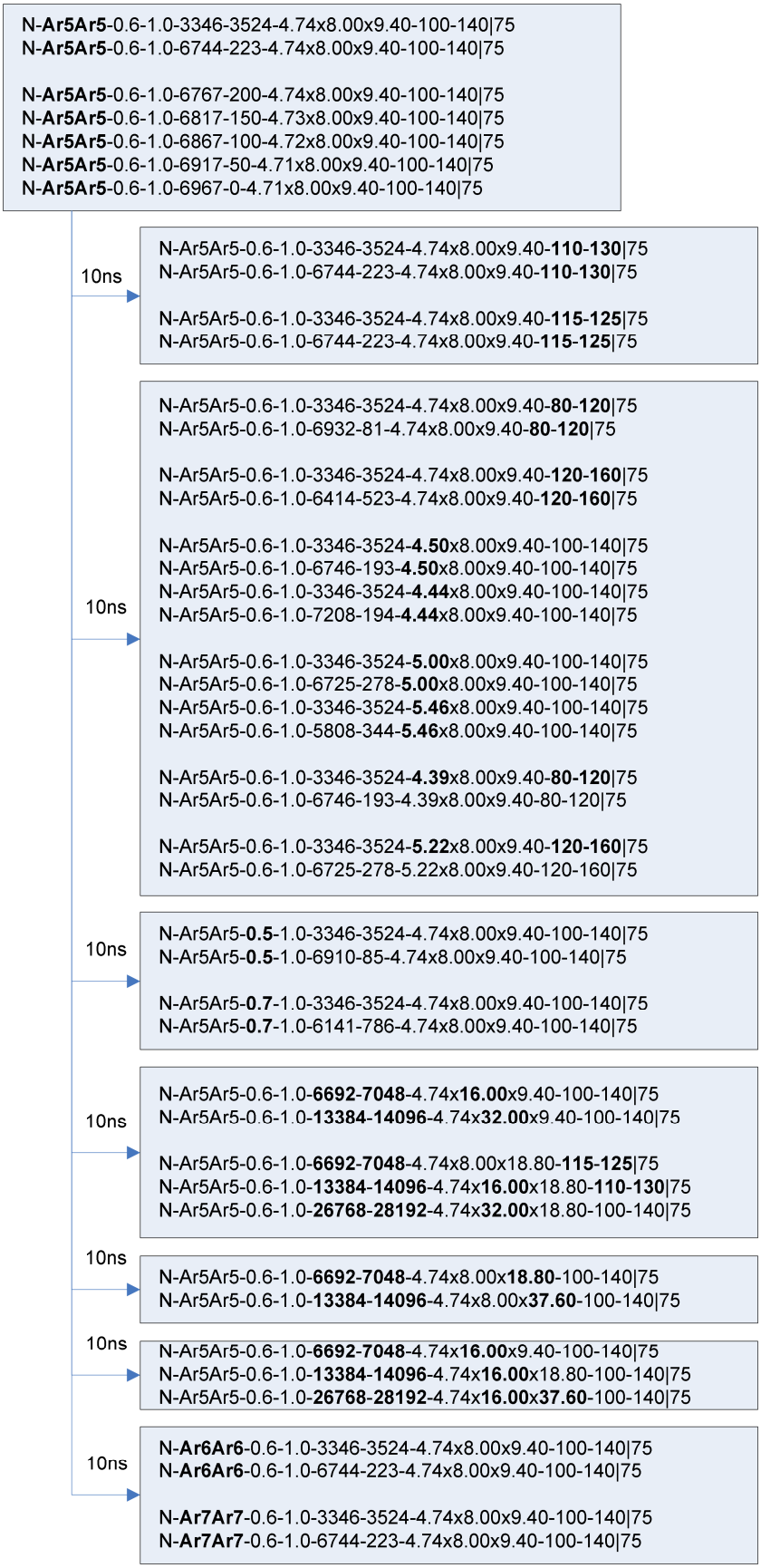
a) EMD simulations of heterophasic interfacial systems



b) EMD simulations of homophasic interfacial systems

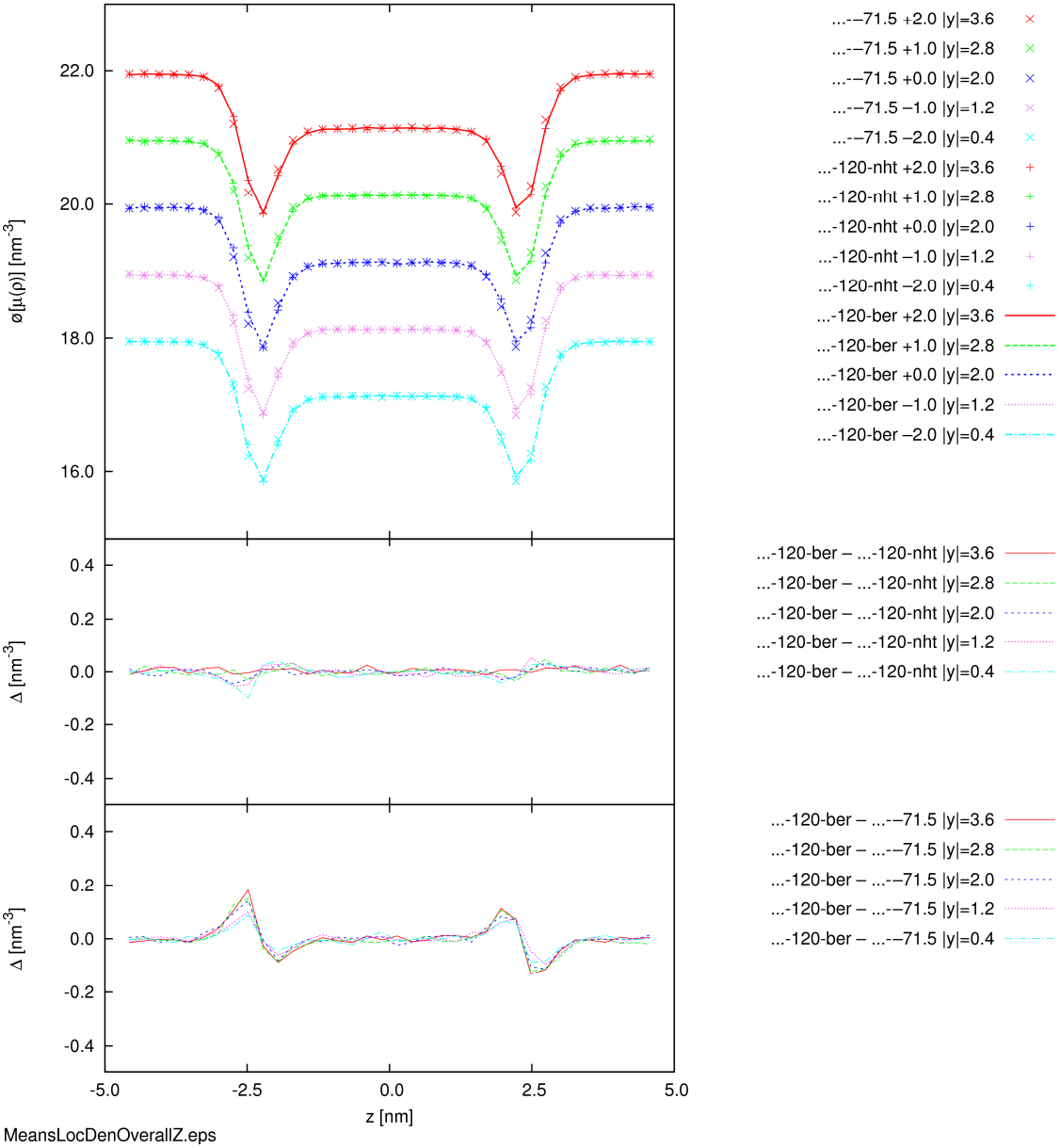


c) NEMD simulations of heterophasic interfacial systems



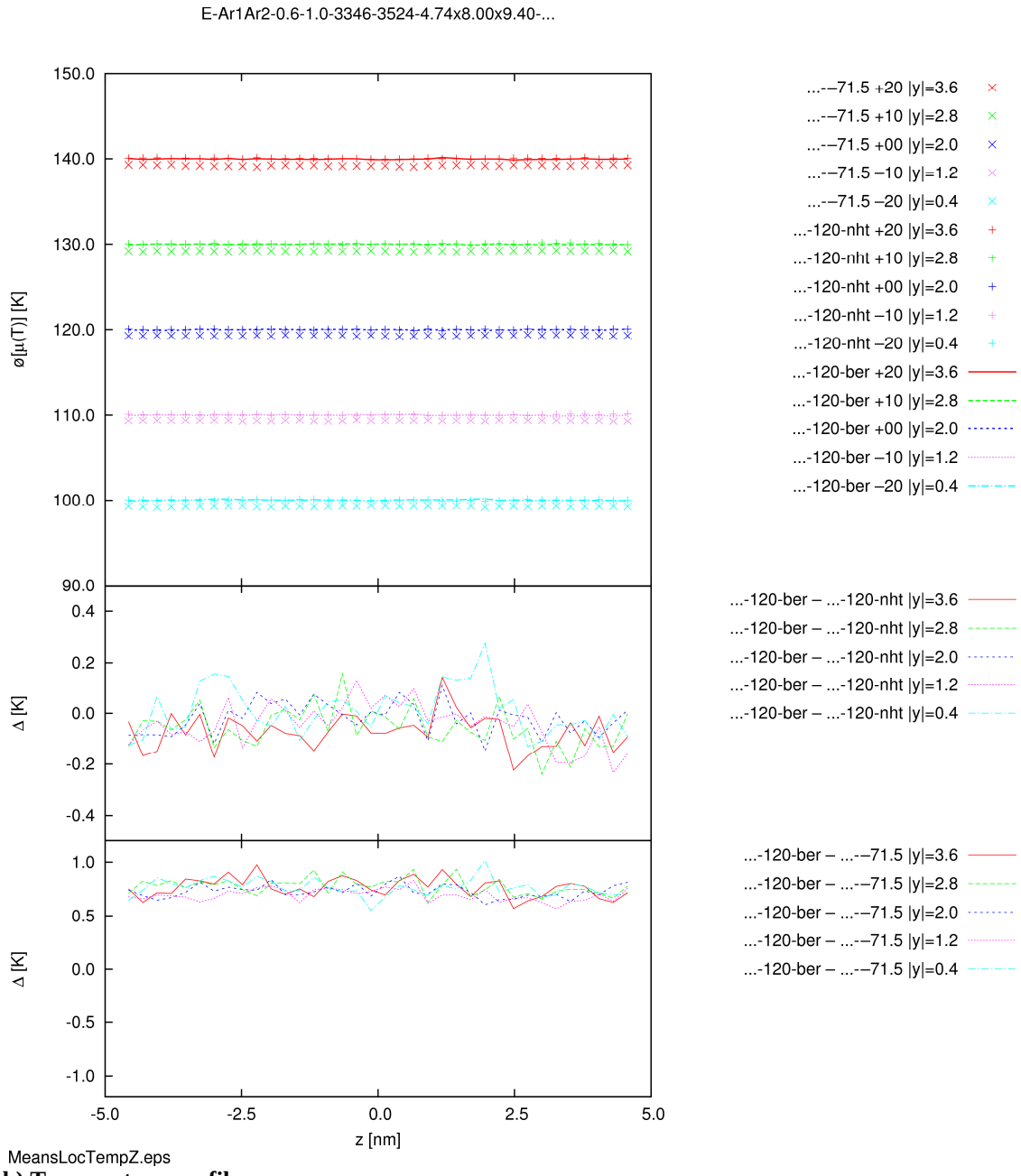
d) NEMD simulations of homophasic interfacial and their corresponding one-phase systems

Figure 22 Production simulations of the different systems in chronological order
We give additionally the lengths of the preparational simulations. Bold symbols indicate the altered simulation parameters.



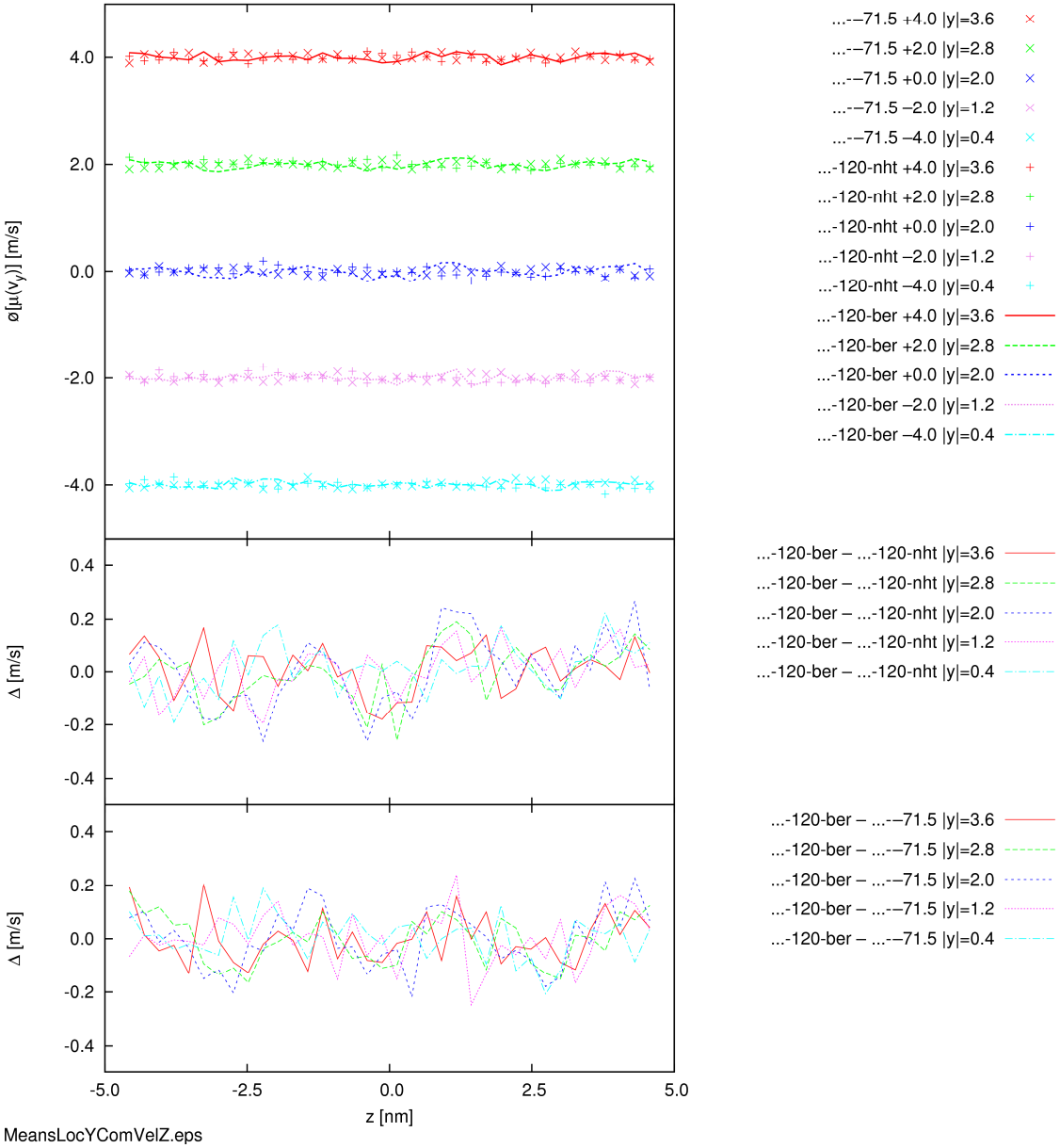
MeansLocDenOverallZ.eps

a) Density profiles



MeansLocTempZ.eps

b) Temperature profiles



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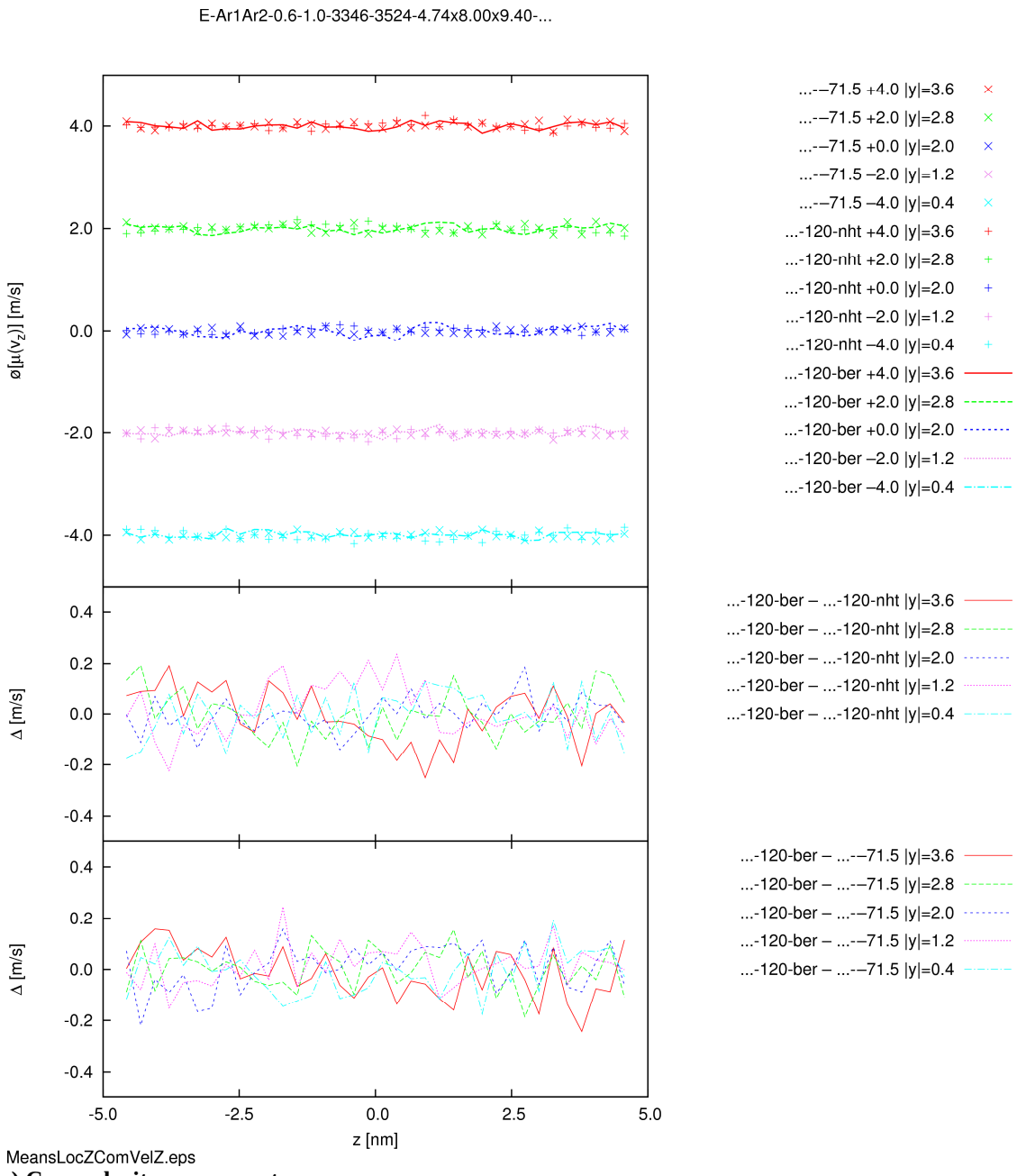


MeansLocZComVelZ.eps

c) Com velocity components

Figure 23 **Comparison of selected local observables in different production simulations of the first heterophasic equilibrium interfacial system E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber, E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-nvt, and E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-−71.5; They start from different initial phase points obtained through the first, second or third preparation route.**

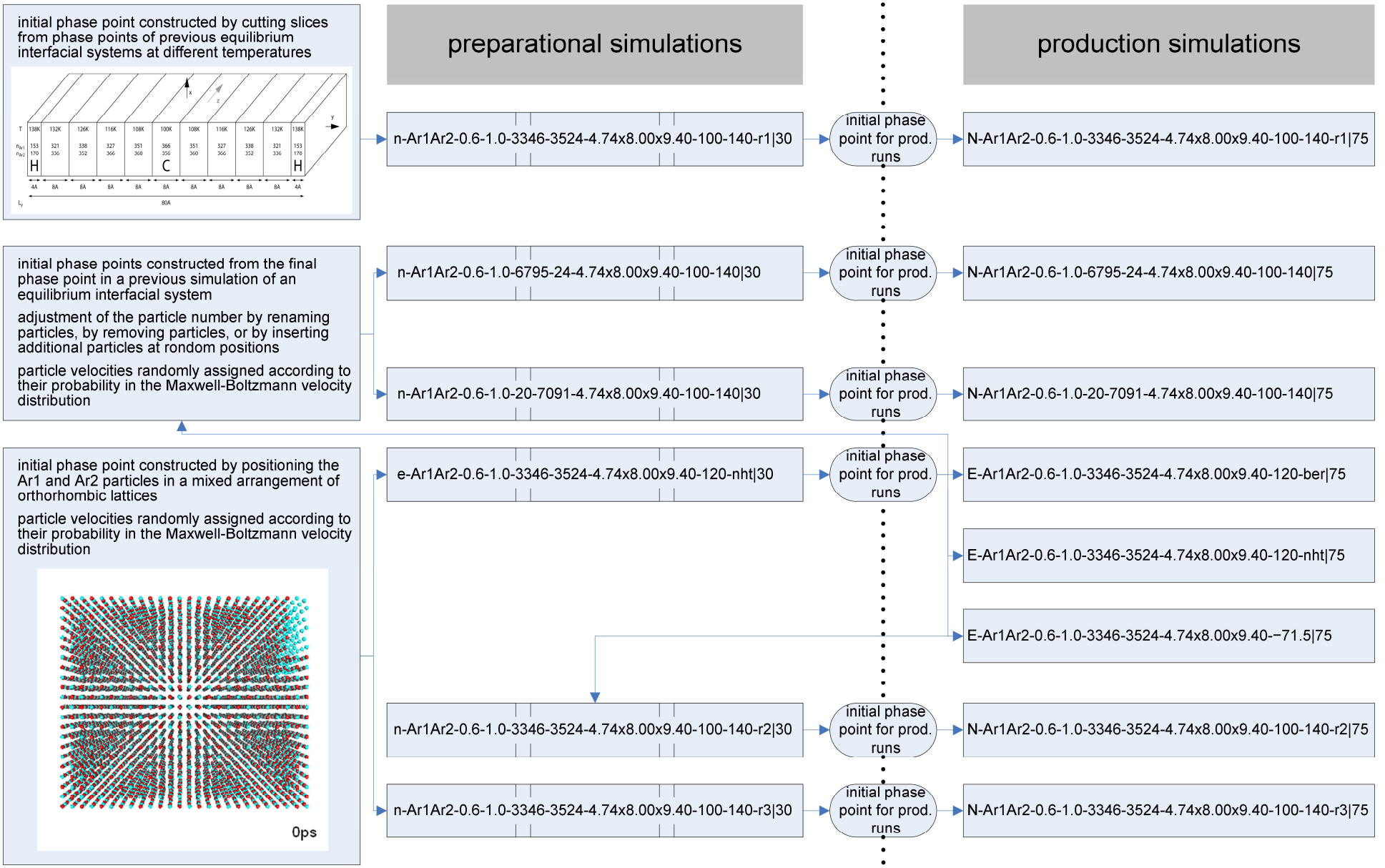
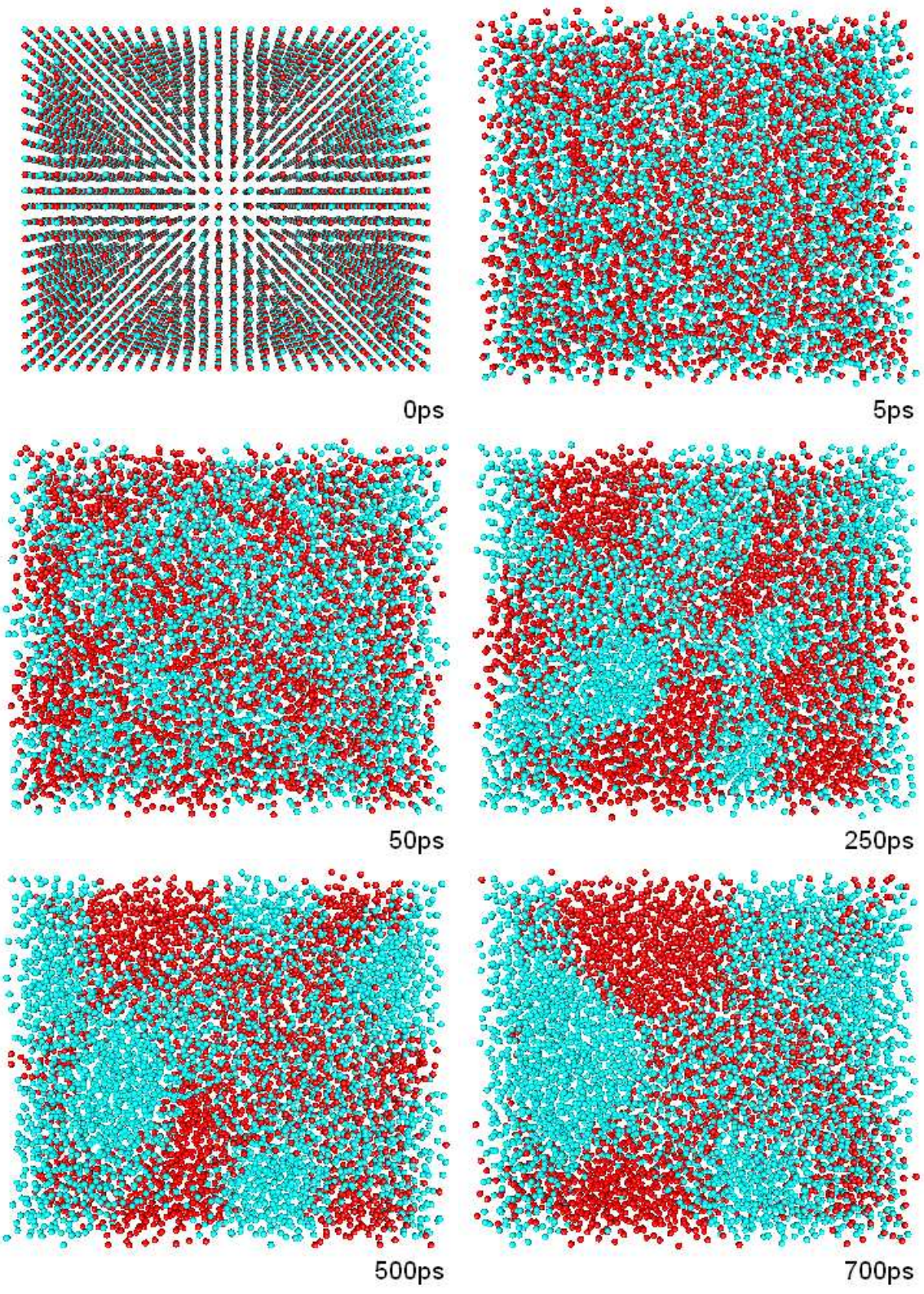


Figure 24 Sequence of the preparational and production simulations of the first systems N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 and E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120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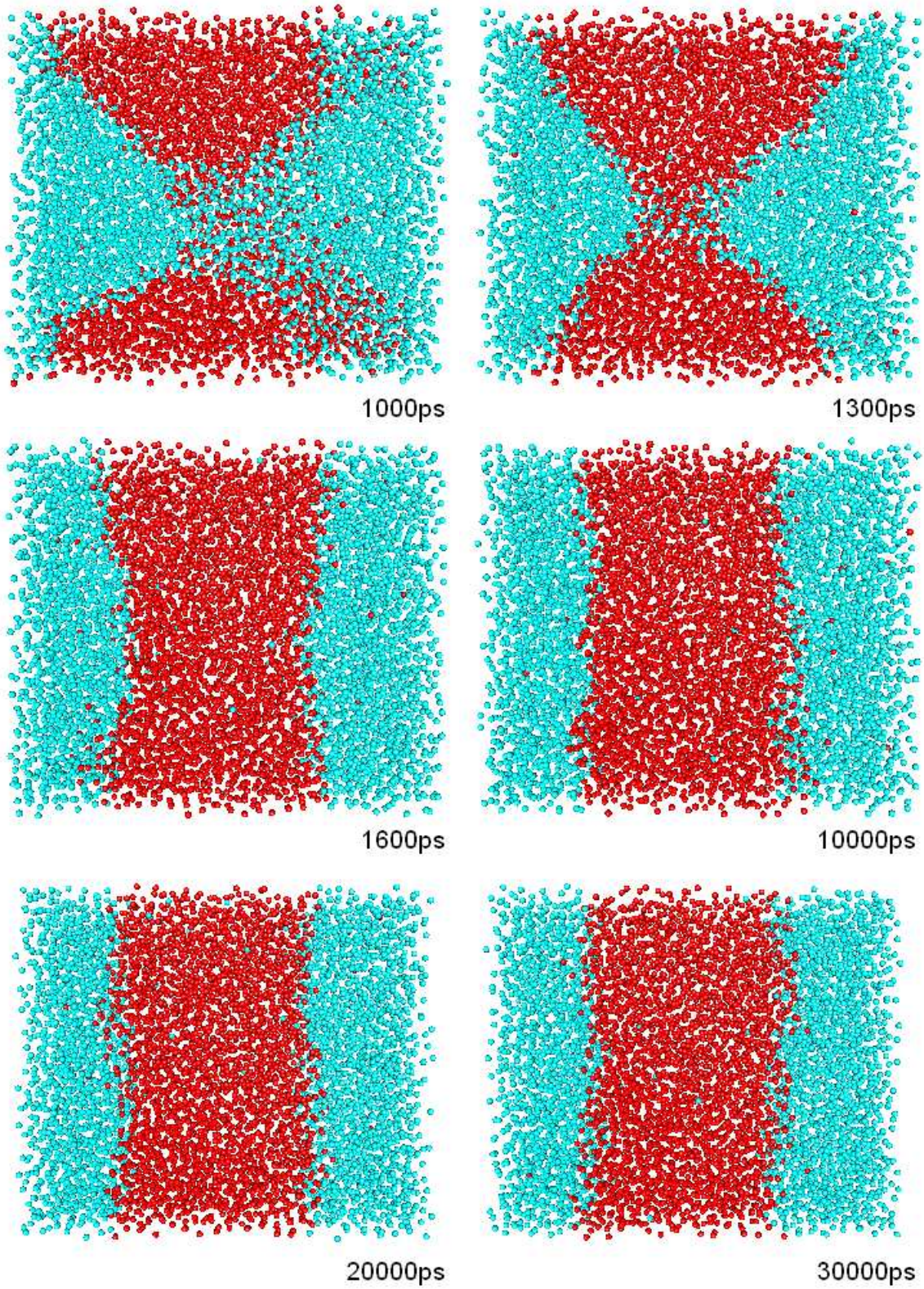
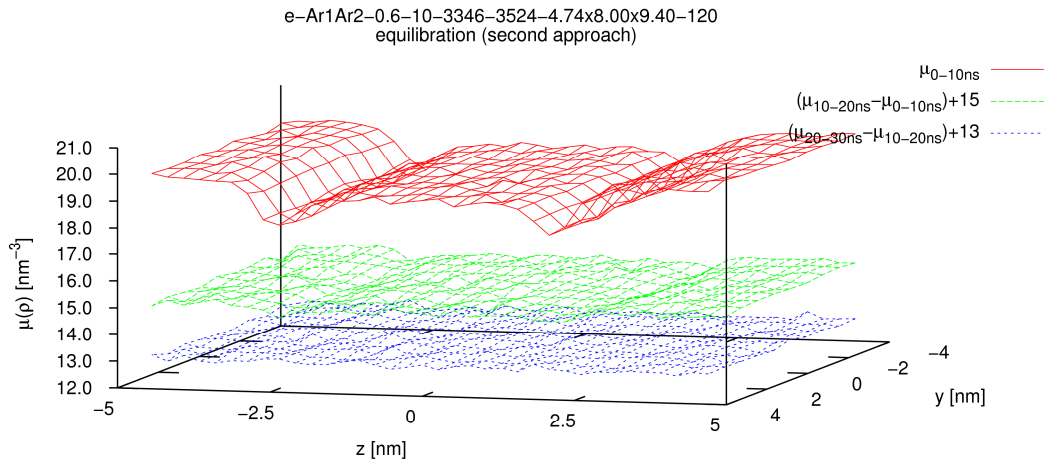
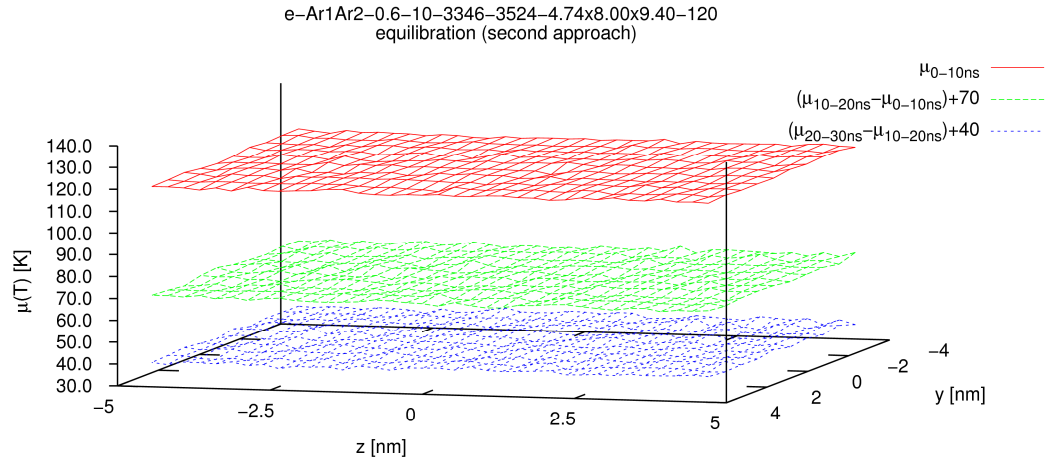


Figure 25 Selected snapshots of the demixing in the equilibration simulation e-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 (second preparation route)



MeansLocDenOverallYZ–diff.eps

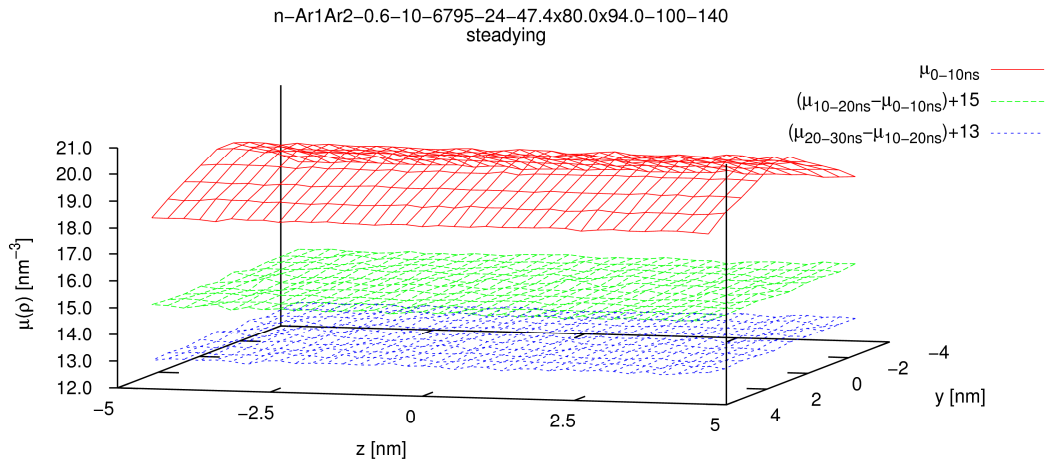
a) Densities



MeansLocTempYZ–diff.eps

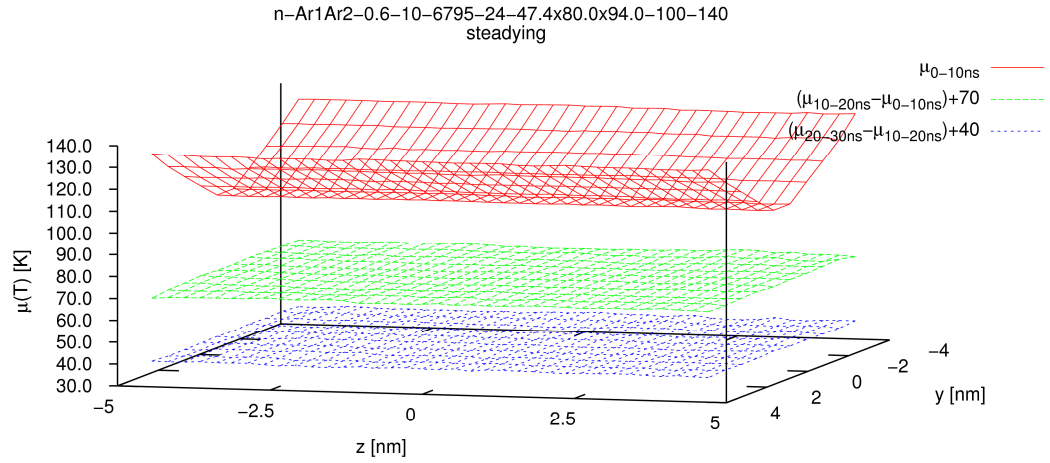
b) Temperatures

Figure 26 Local densities determined over three 10ns long successive segments of the equilibration simulation e-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 (second preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.



MeansLocDenOverallYZ–diff.eps

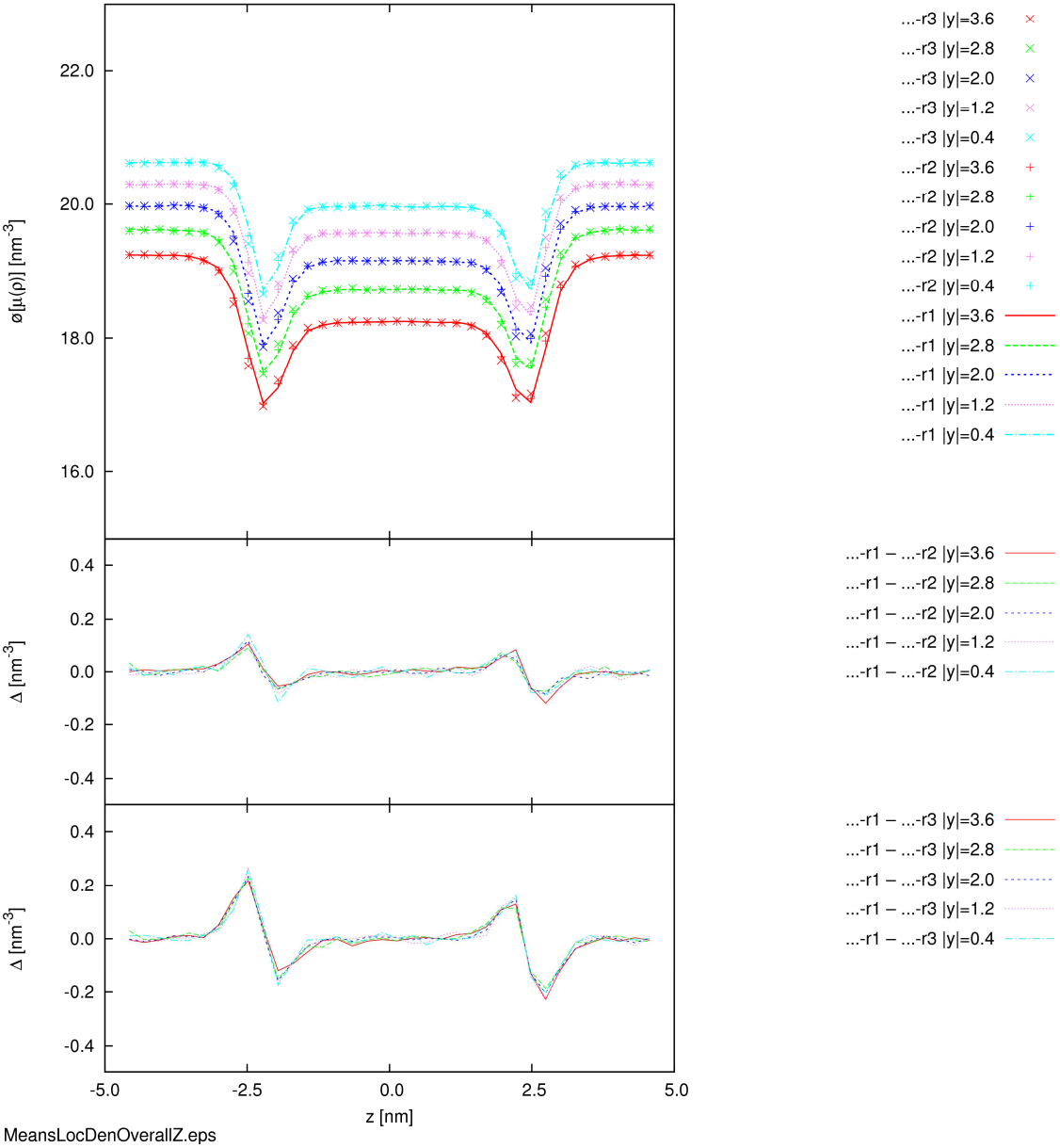
a) Densities



MeansLocTempYZ–diff.eps

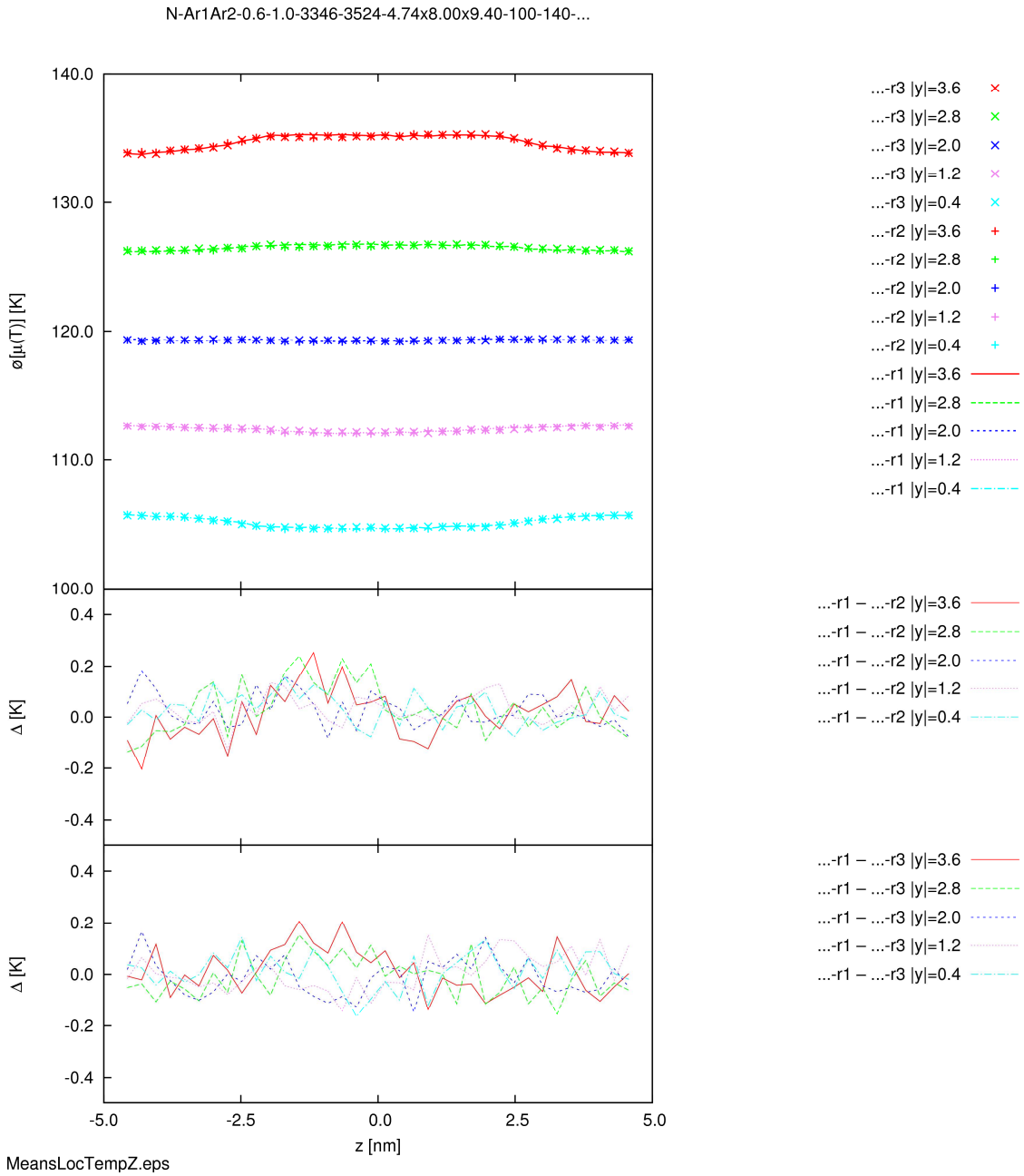
b) Temperatures

Figure 27 Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.



MeansLocDenOverallZ.eps

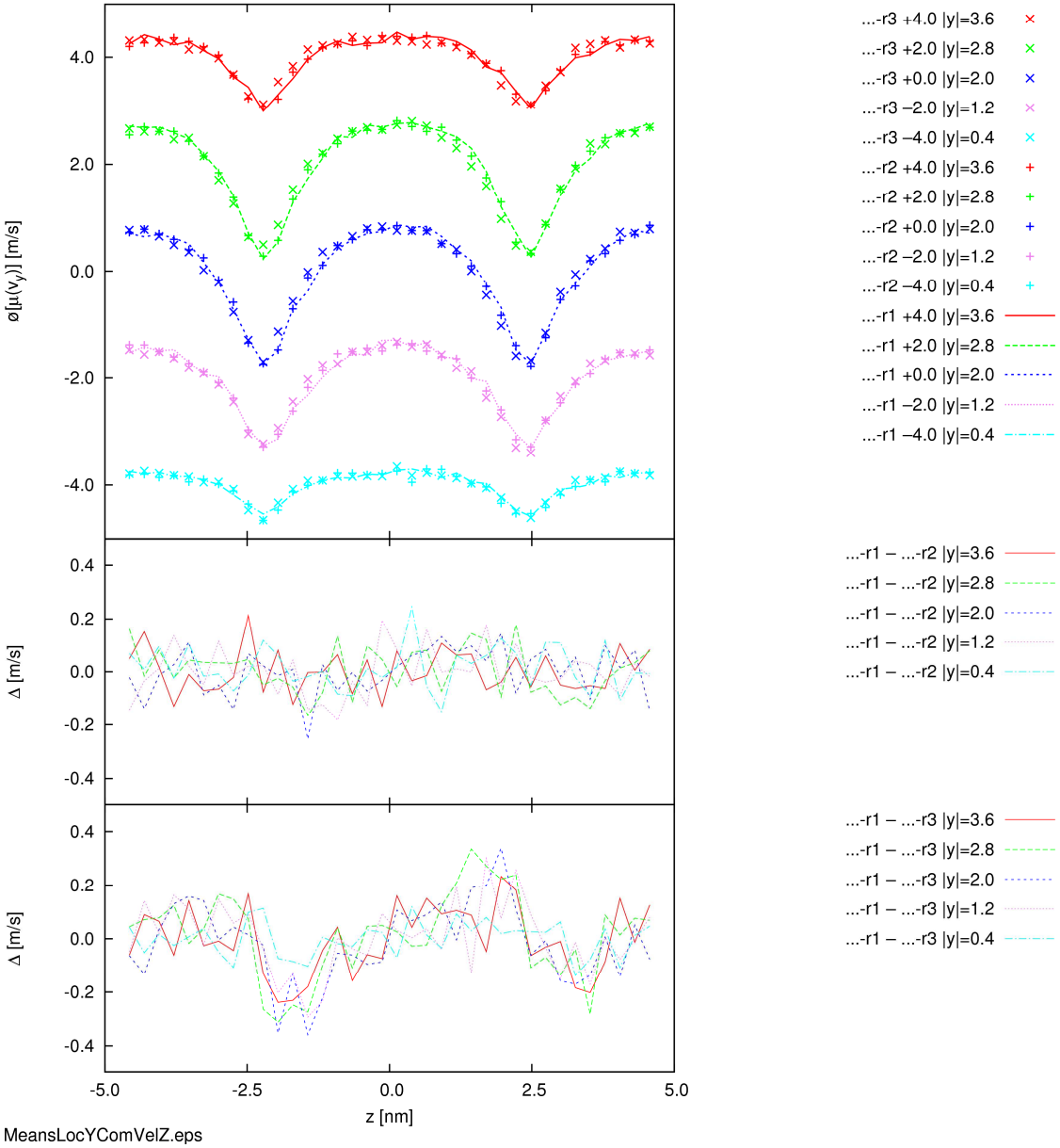
a) Density profiles



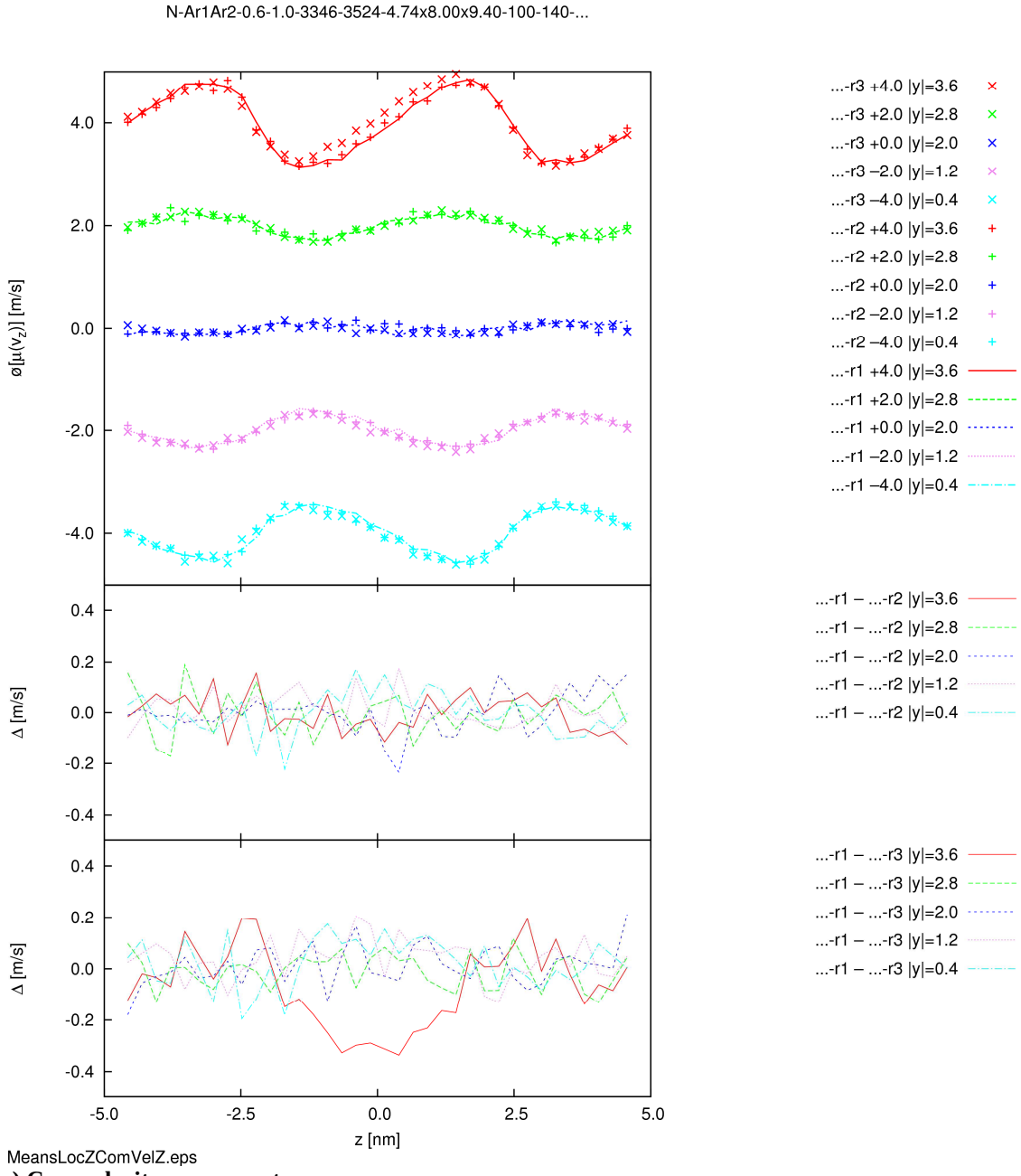
MeansLocTempZ.eps

b) Temperature profiles

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-...



MeansLocYComVelZ.eps



MeansLocZComVelZ.eps

c) Com velocity components

Figure 28 **Comparison of selected local observables in separate production simulations of the first heterophasic nonequilibrium interfacial system N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1, N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r2, and N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r3 They start from different initial phase points obtained through the first, second or third preparation route.**

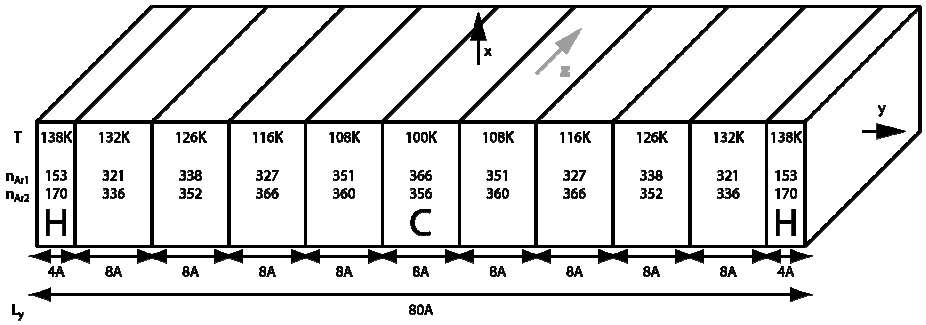
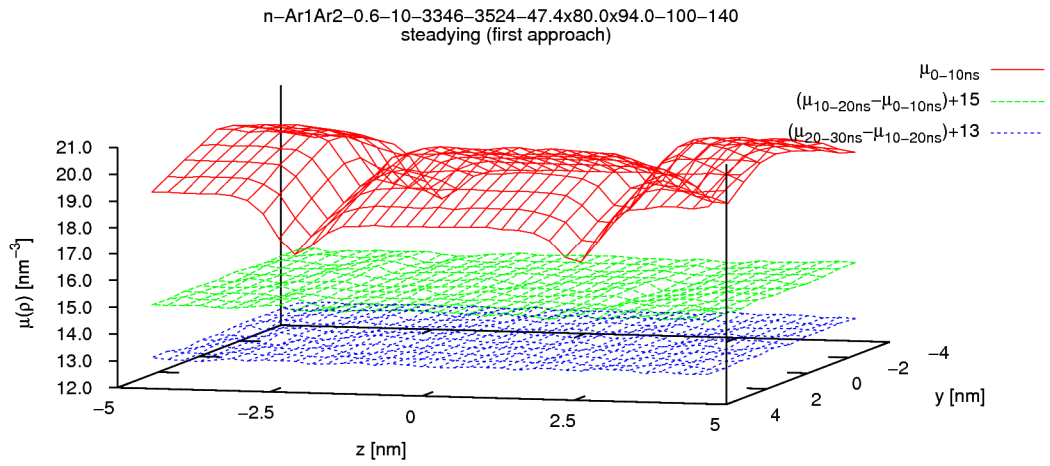
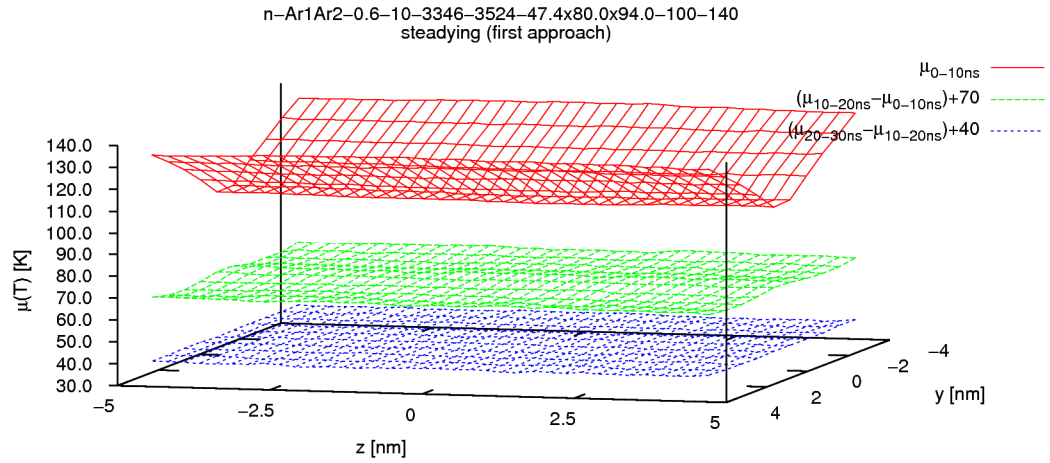


Figure 29 Preparation of an initial phase point from cut-outs of phase points in Buhn's NPT simulations
We link the different cut-outs together in *y*-direction to obtain a phase point that resembles one to be found in the first nonequilibrium interfacial system. The figure shows additionally the numbers of particles found in the different cut-outs.



MeansLocDenOverallYZ–diff.eps

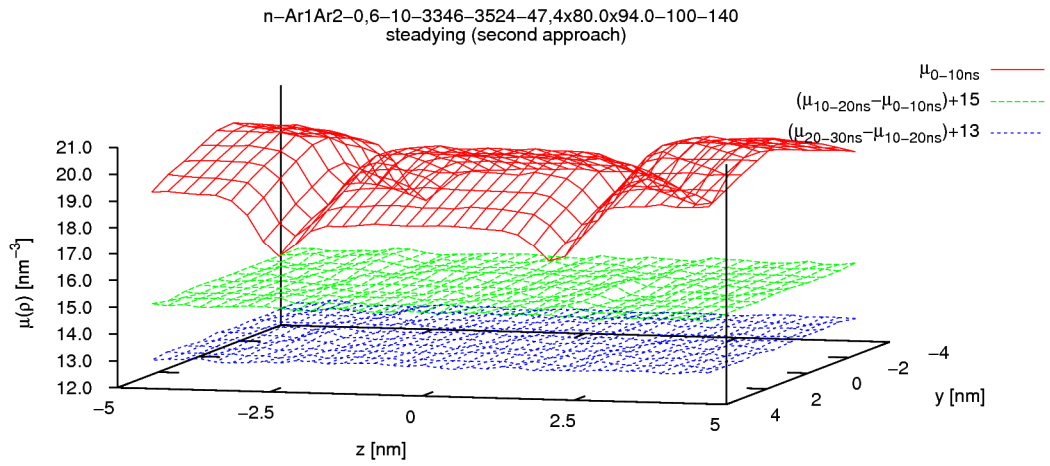
a) Densities



MeansLocTempYZ–diff.eps

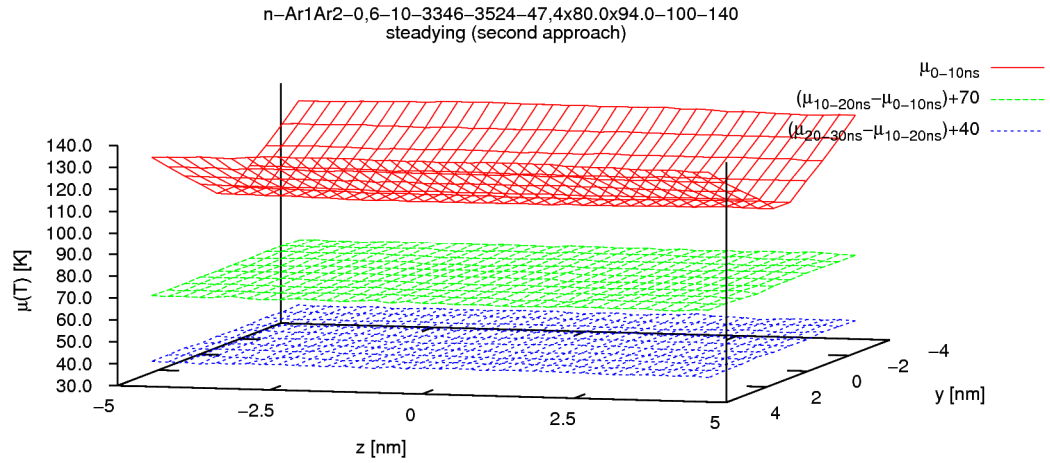
b) Temperatures

Figure 30 Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (first preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.



MeansLocDenOverallYZ–diff.eps

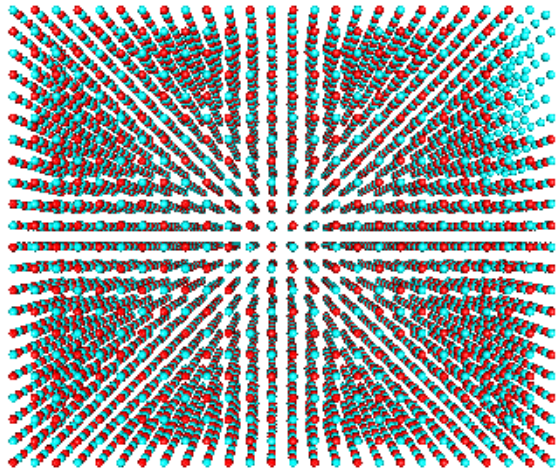
a) Densities



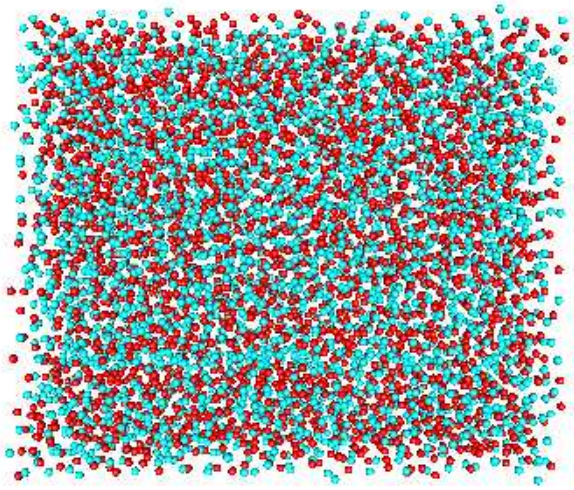
MeansLocTempYZ–diff.eps

b) Temperatures

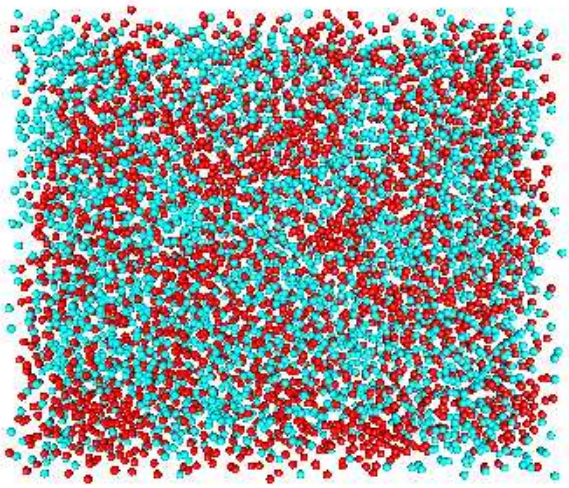
Figure 31 Local observables determined over three 10ns long successive segments of the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (second preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.



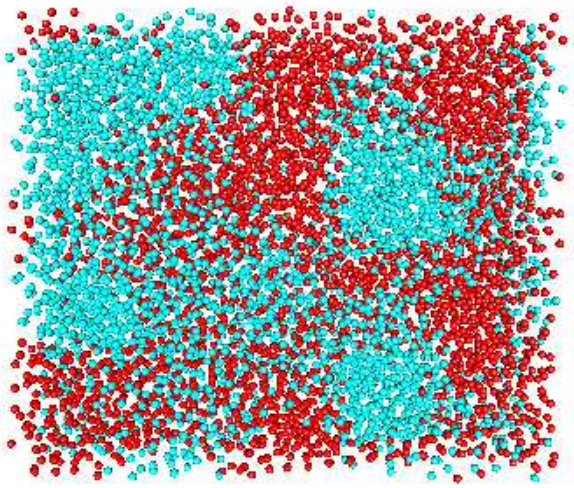
0ps



5ps



50ps



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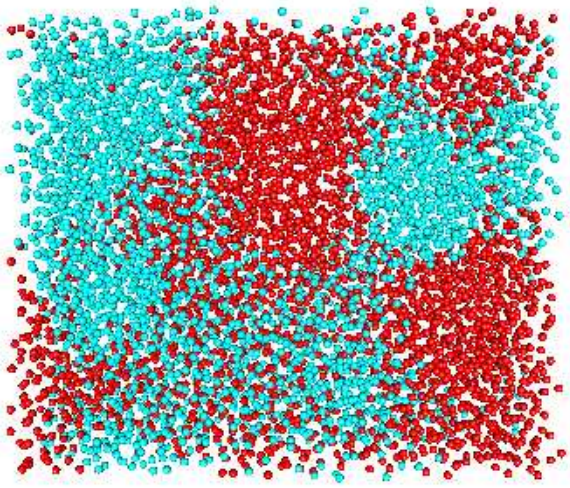
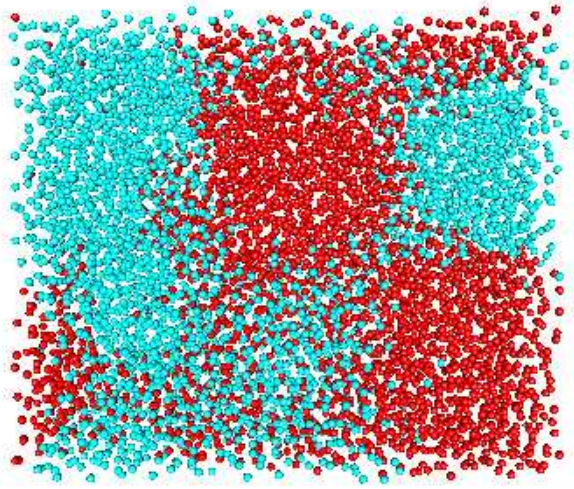


400ps



500ps

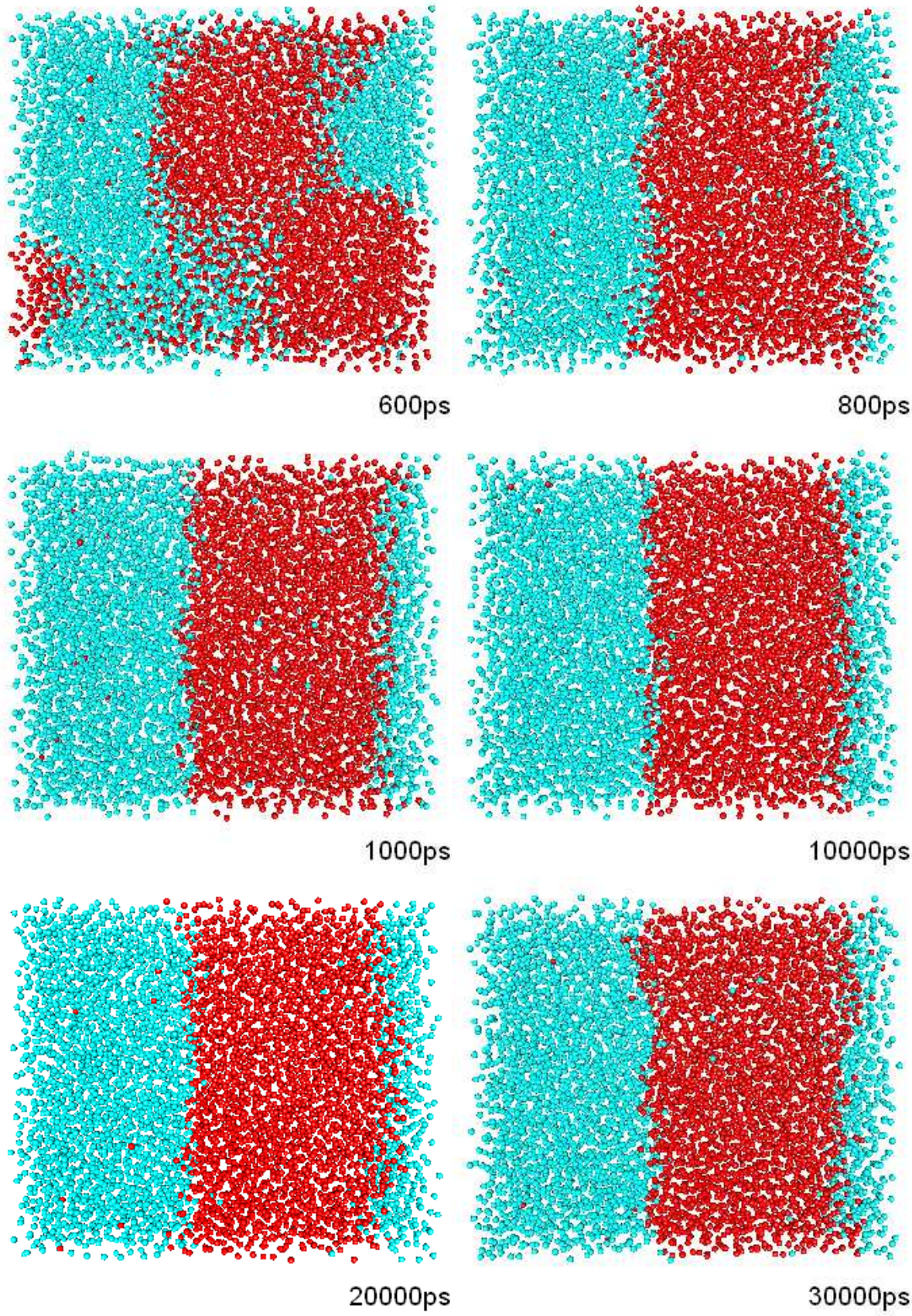
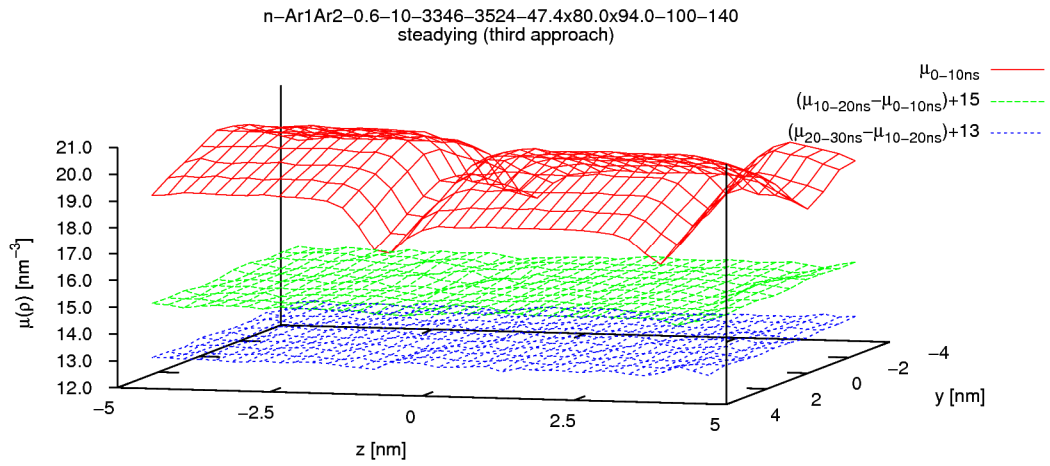
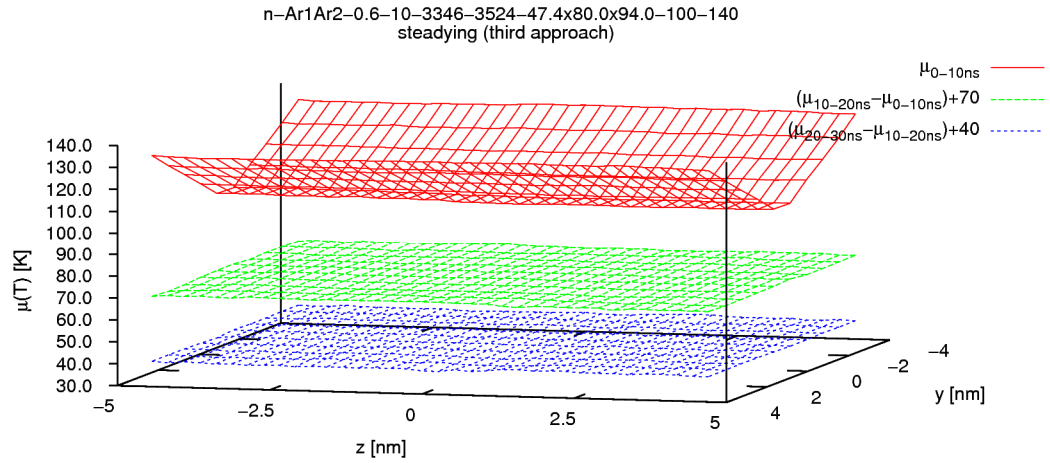


Figure 32 Selected snapshots of the demixing in the steadying simulation n-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (third preparation route)



MeansLocDenOverallYZ–diff.eps

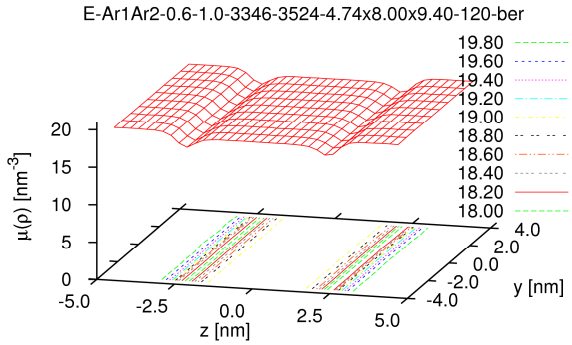
a) Densities



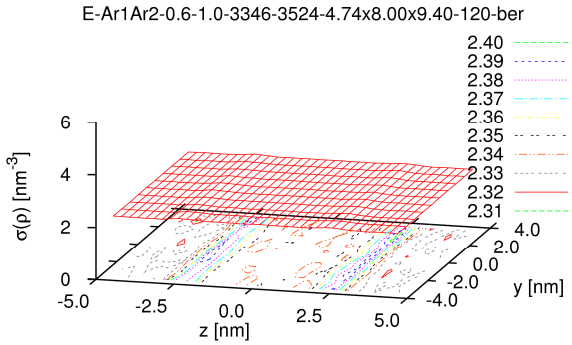
MeansLocTempYZ–diff.eps

b) Temperatures

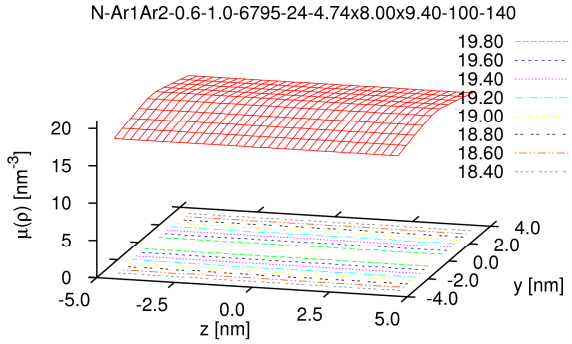
Figure 33 Local observables determined over three 10ns long successive segments of the steadying simulation N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 (third preparation route)
Note that only the changes from the first to the second and from the second to the third segment are displayed for better readability.



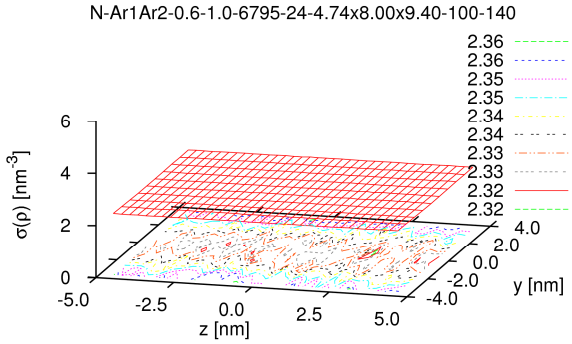
MeansLocDenOverallYZ.eps



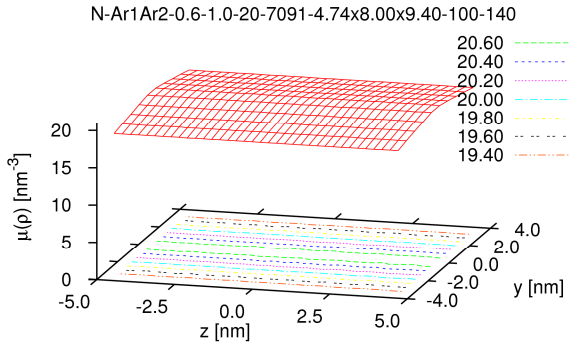
SigmaLocDenOverallYZ.eps



MeansLocDenOverallYZ.eps



SigmaLocDenOverallYZ.eps



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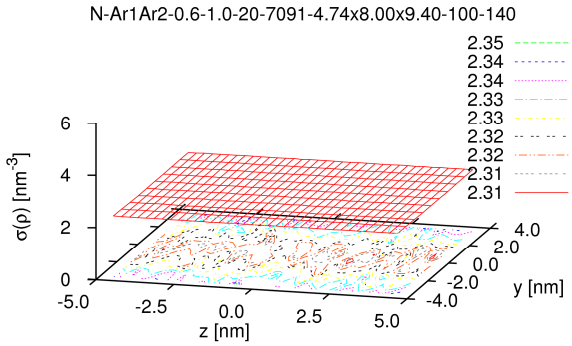

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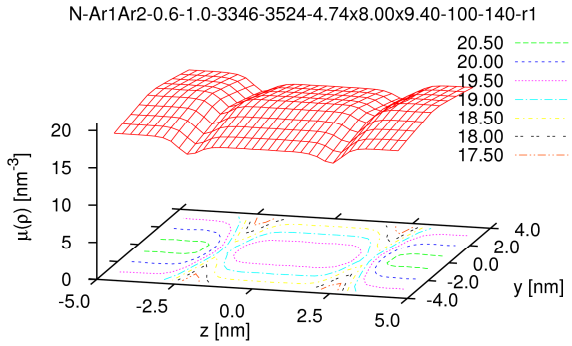

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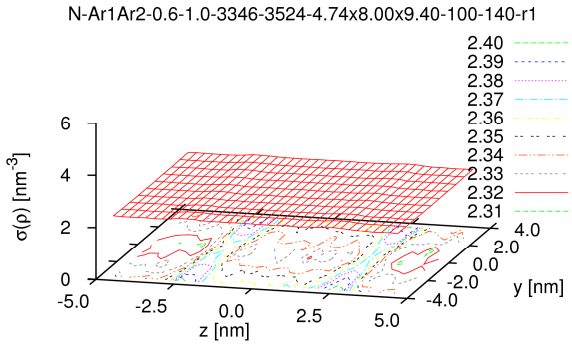
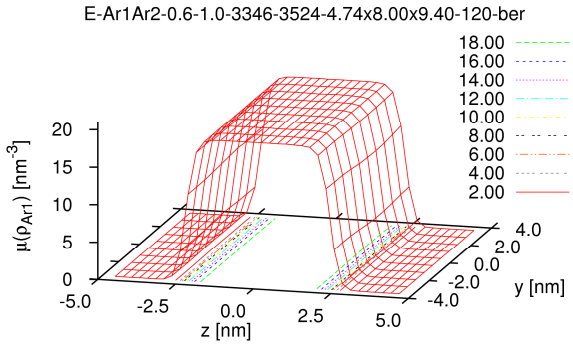


SigmaLocDenOverallYZ.eps

a) Densities



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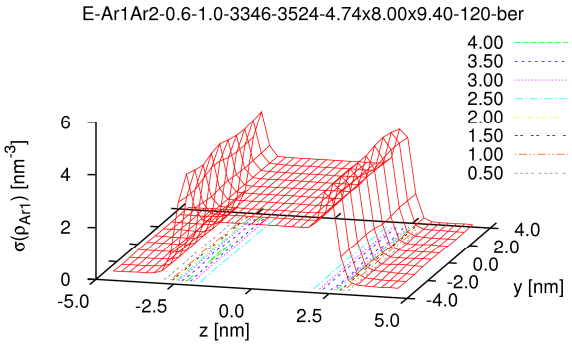

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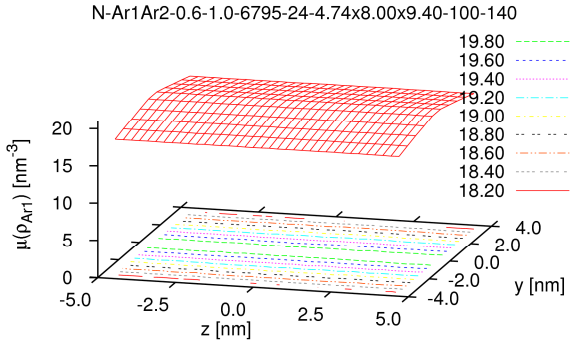

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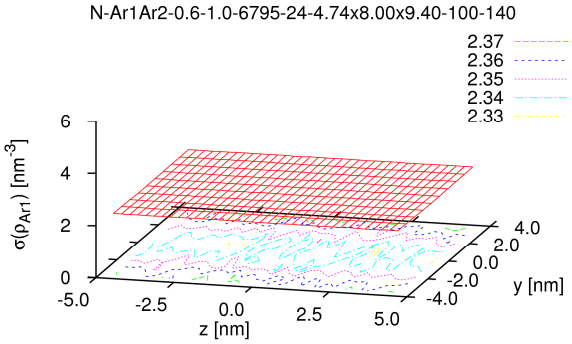

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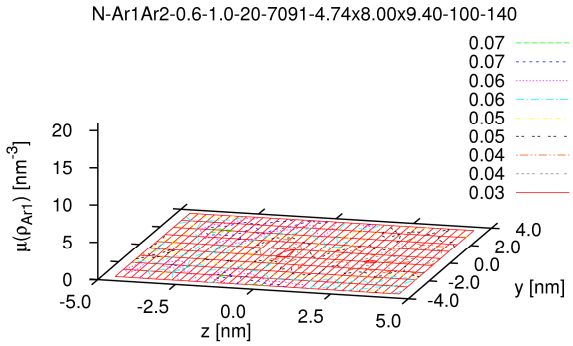
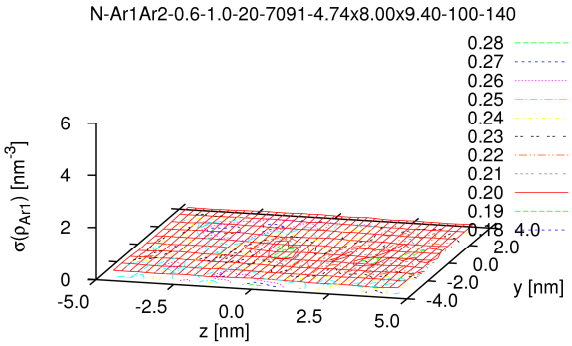
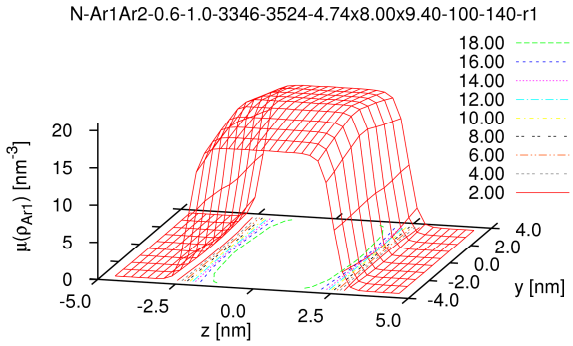


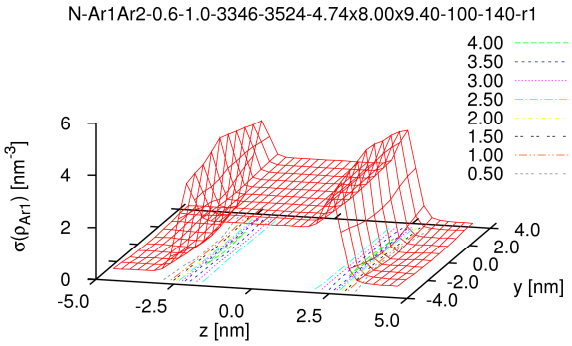
MeansLocDenAr1YZ.eps



SigmaLocDenAr1YZ.eps

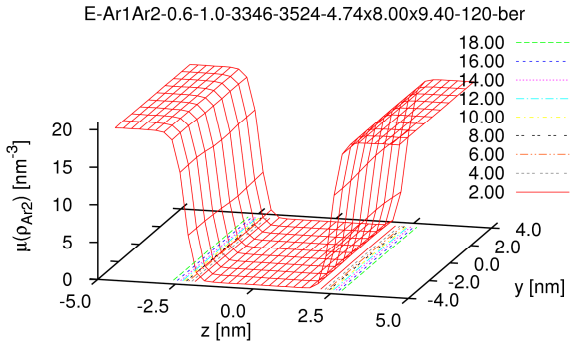


MeansLocDenAr1YZ.eps

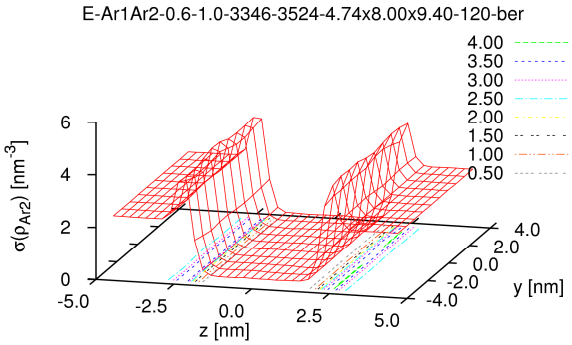


SigmaLocDenAr1YZ.eps

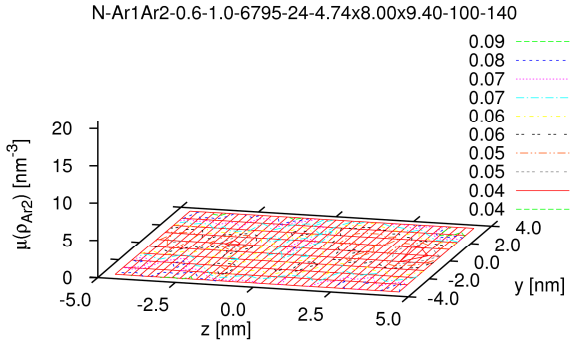
b) ArA partial densities



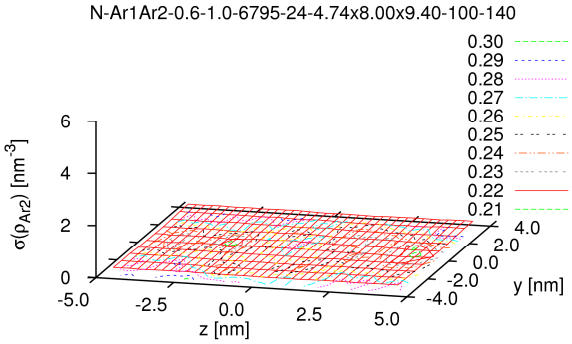
MeansLocDenAr2YZ.eps



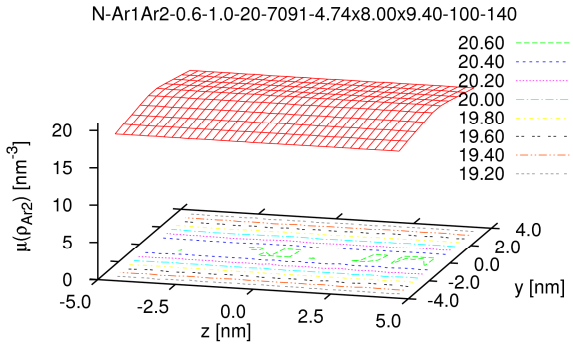
SigmaLocDenAr2YZ.eps



MeansLocDenAr2YZ.eps



SigmaLocDenAr2YZ.eps



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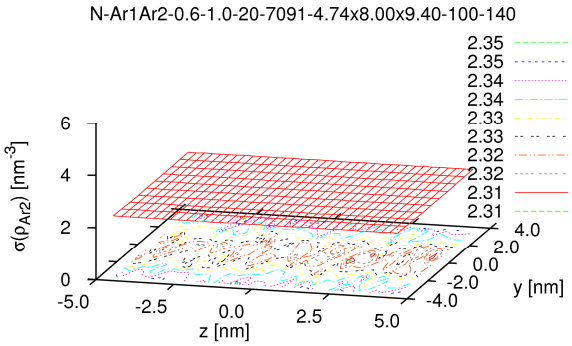

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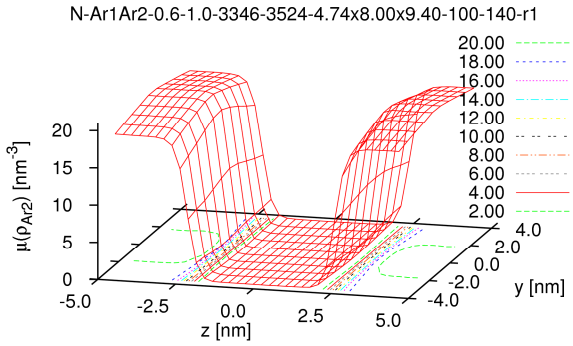

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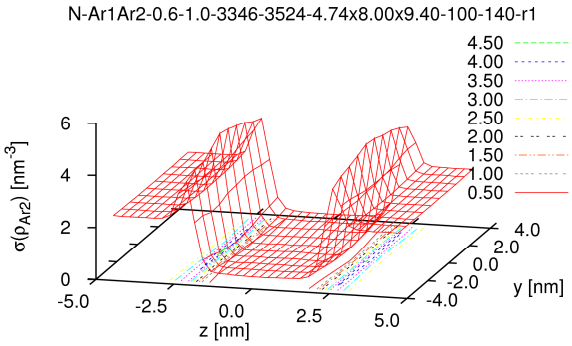
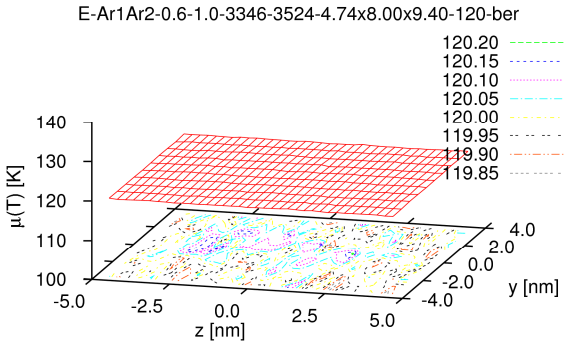
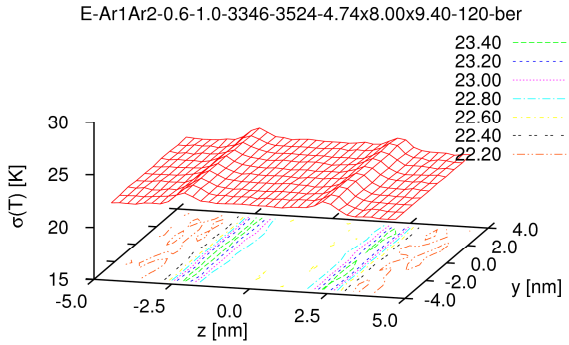


SigmaLocDenAr2YZ.eps

c) ArB partial densities



MeansLocTempYZ.eps



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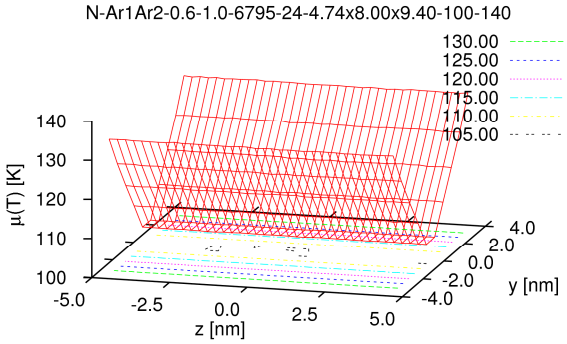

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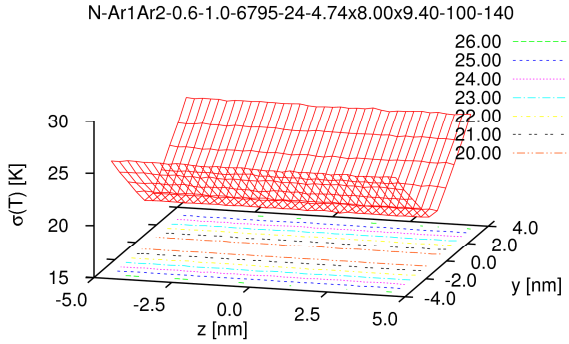
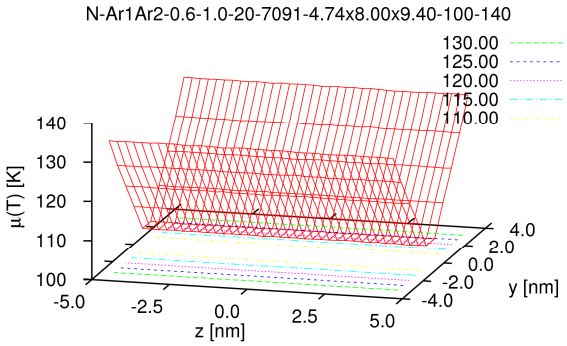


SigmaLocTempYZ.eps



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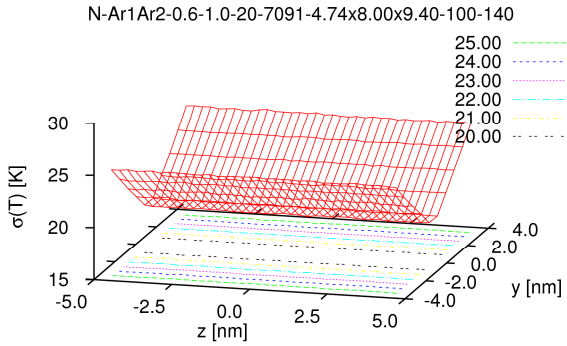
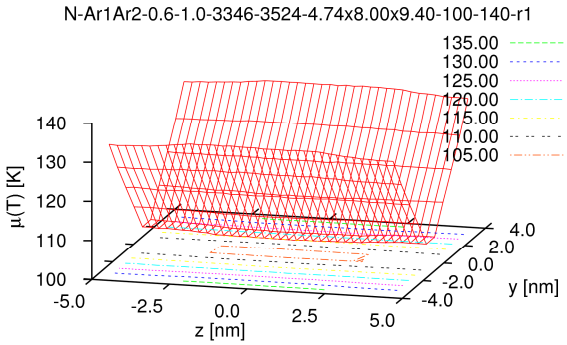
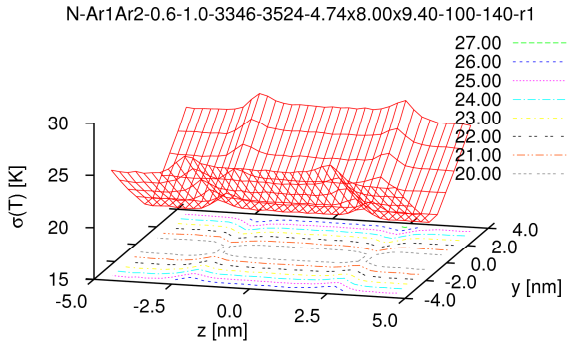


SigmaLocTempYZ.eps

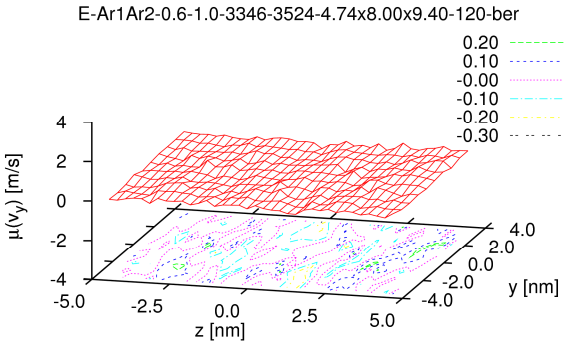


MeansLocTempYZ.eps

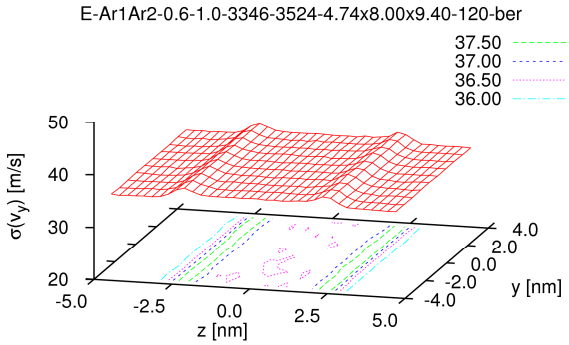


SigmaLocTempYZ.eps

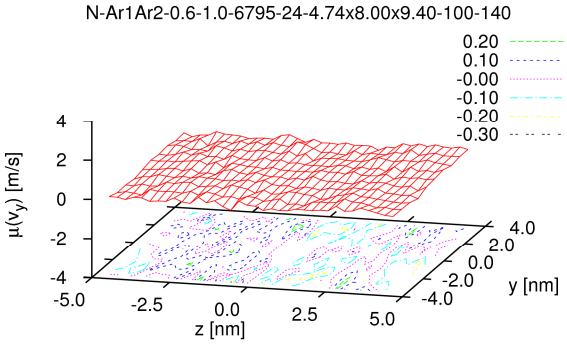
d) Temperatures



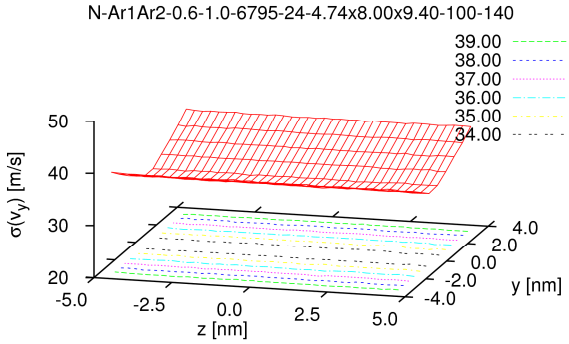
MeansLocYComVelYZ.eps



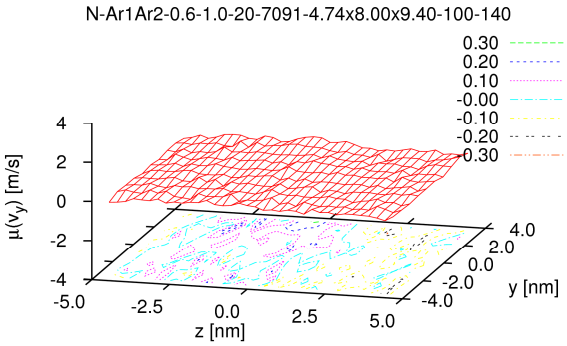
SigmaLocYComVelYZ.eps



MeansLocYComVelYZ.eps



SigmaLocYComVelYZ.eps



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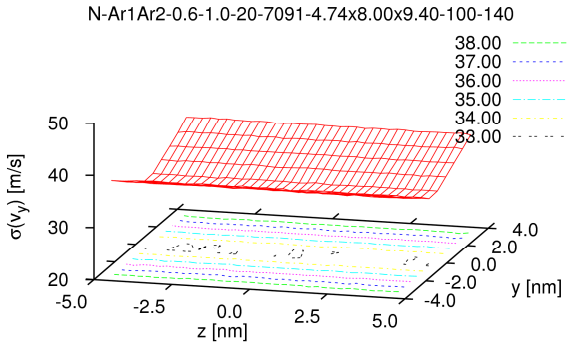

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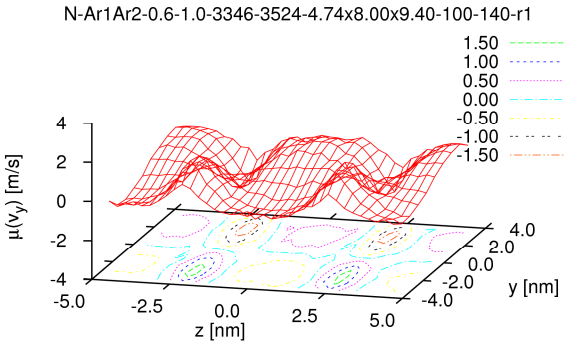

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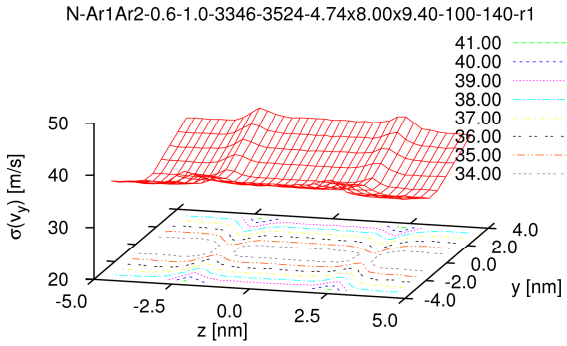

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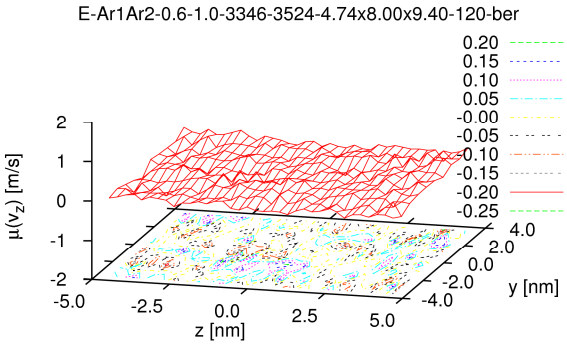
e) Com *y*-velocities



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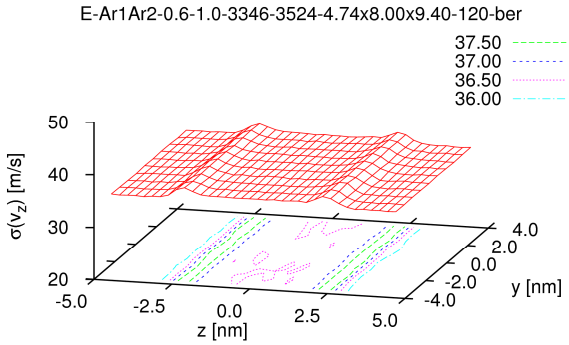

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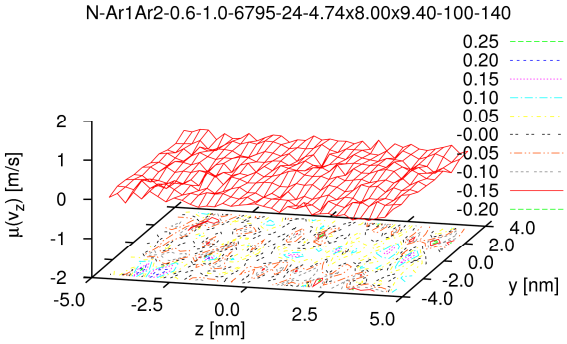

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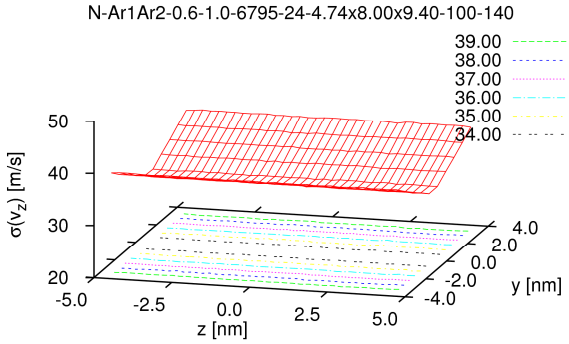

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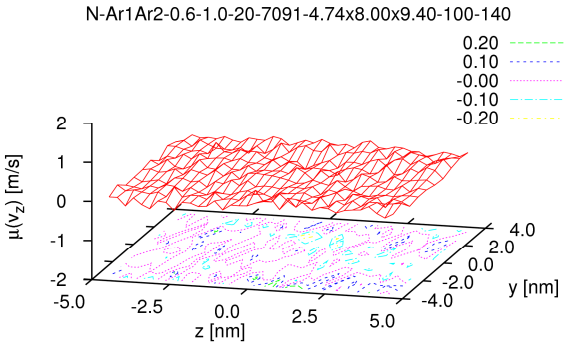

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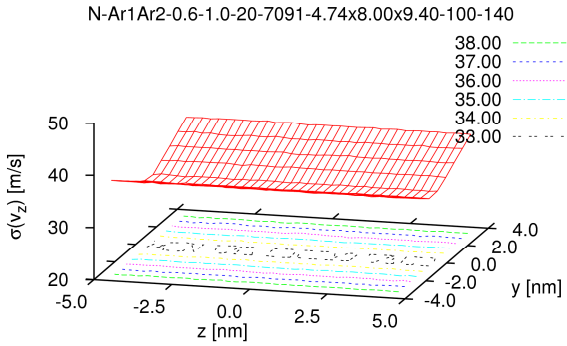

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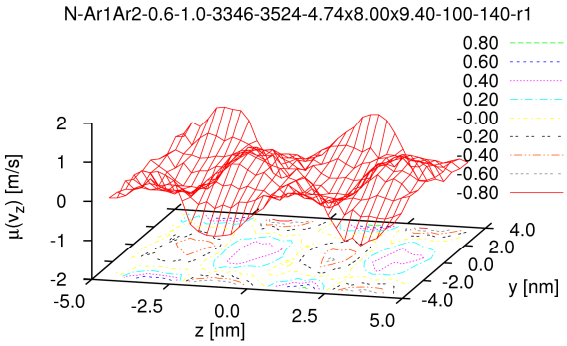

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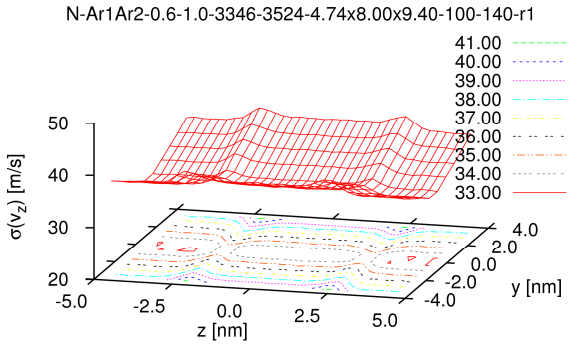
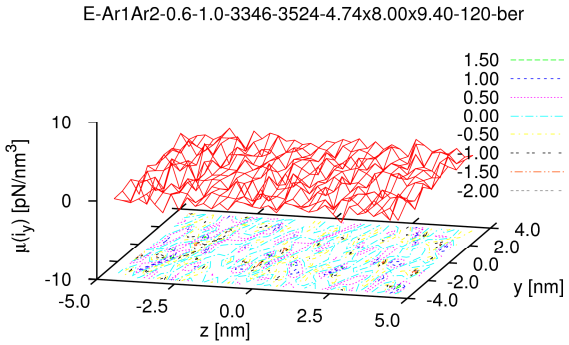


SigmaLocZComVelYZ.eps

f) Com *z*-velocities



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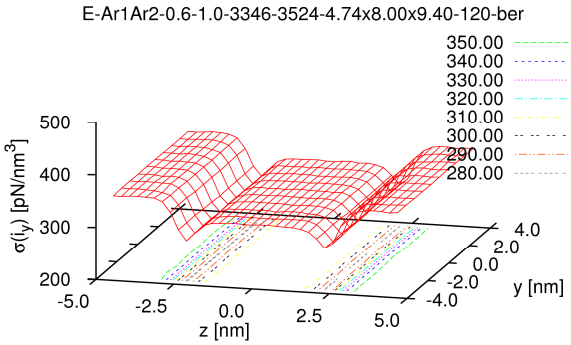

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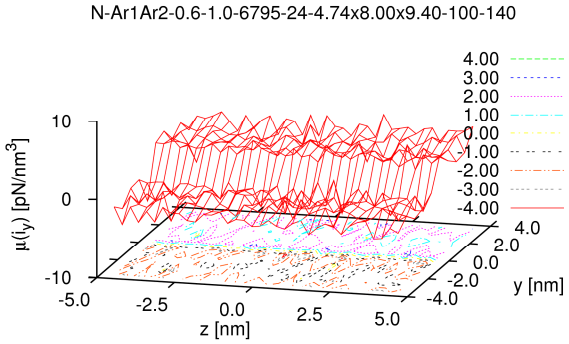

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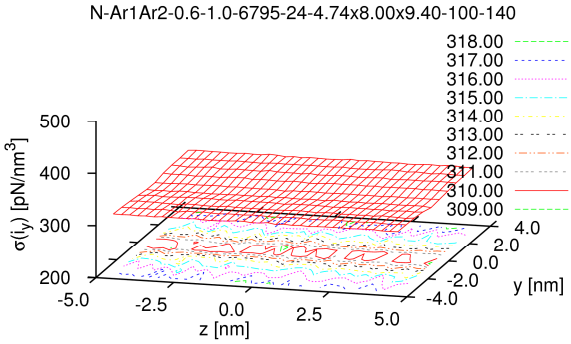

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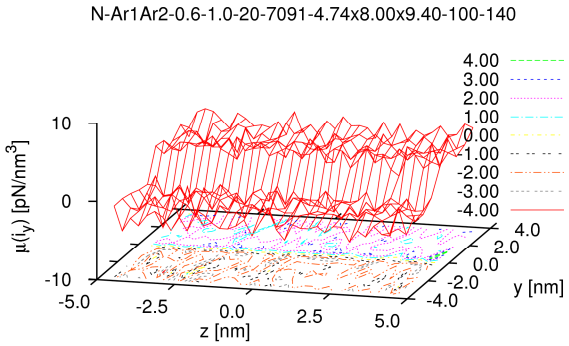

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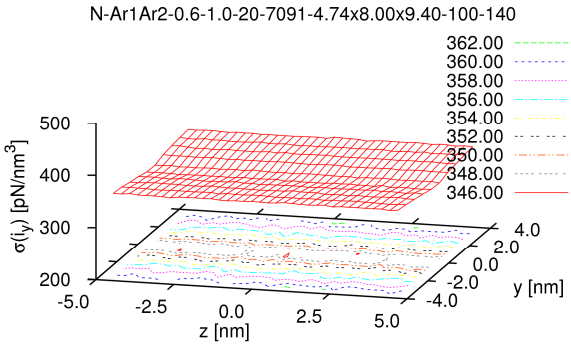

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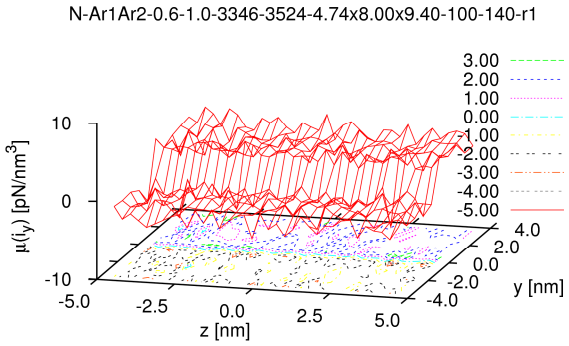

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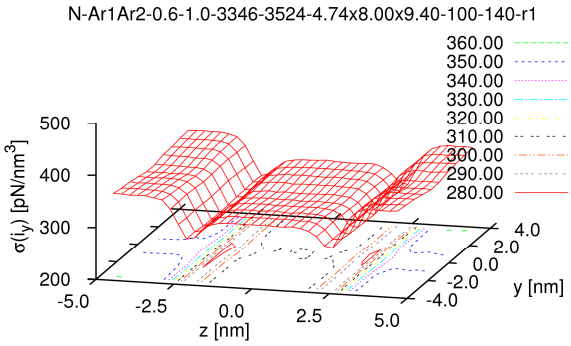
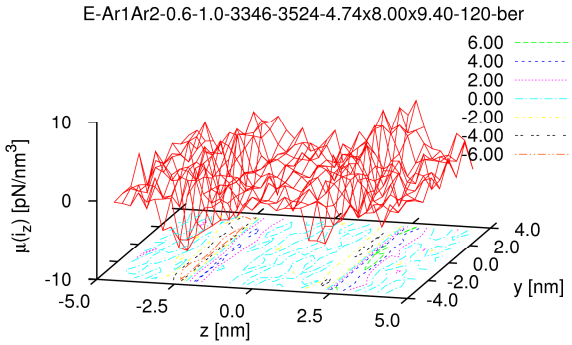


SigmaLocYVolForceYZ.eps

g) Interatomic volume *y*-forces



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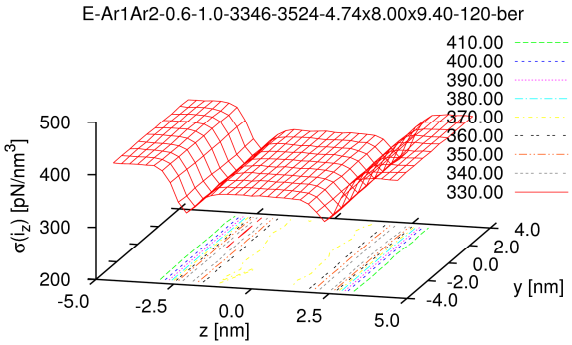

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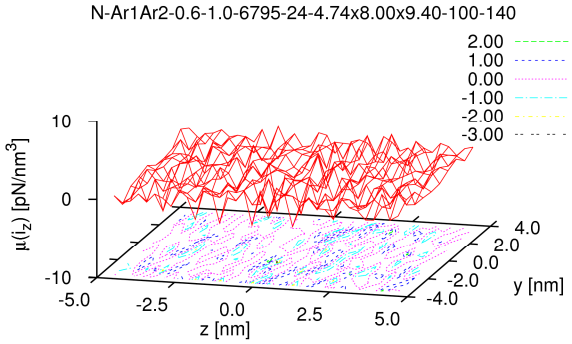

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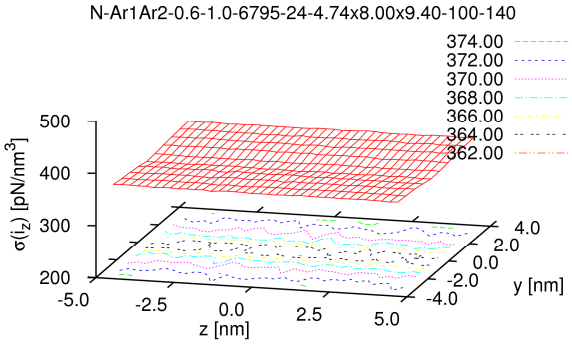

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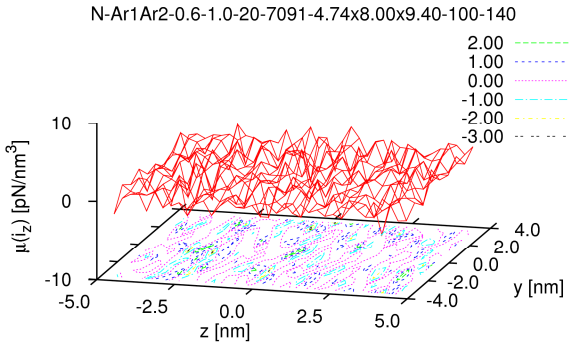

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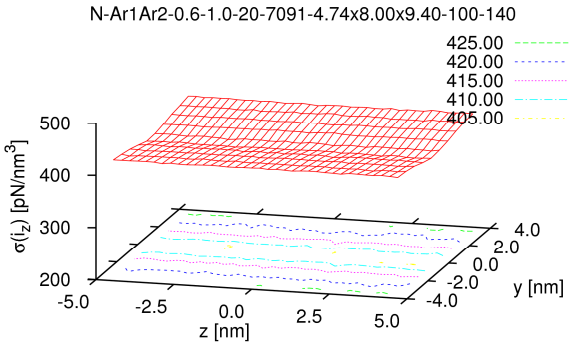

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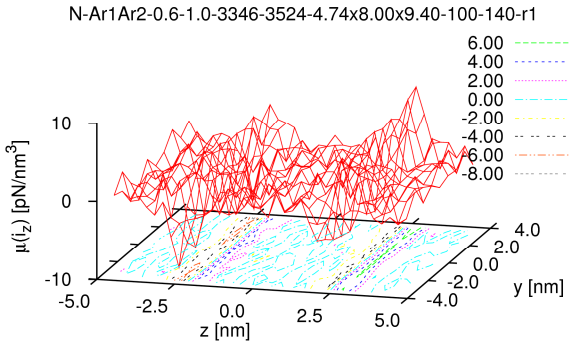

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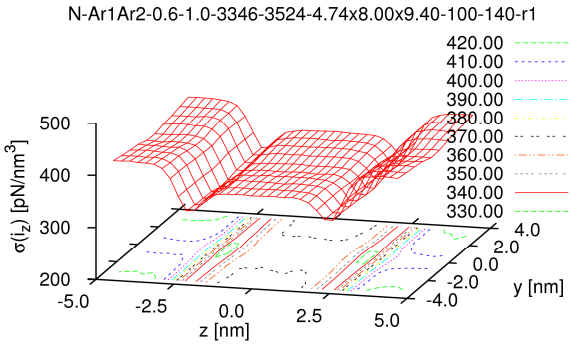
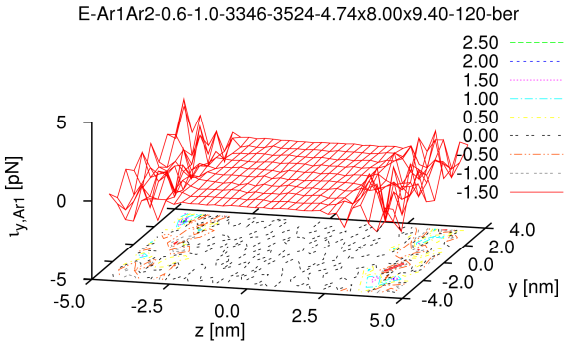
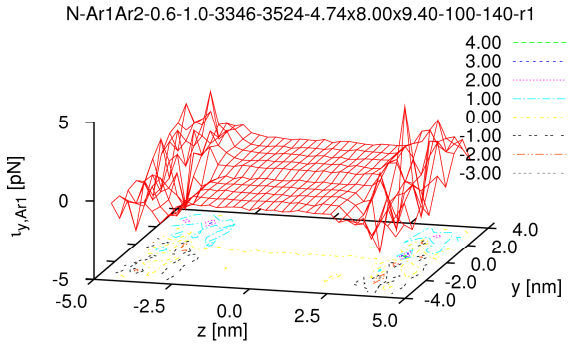


SigmaLocZVolForceYZ.eps

h) Interatomic volume *z*-forces



MeansLocYPartForceAr1YZ.eps



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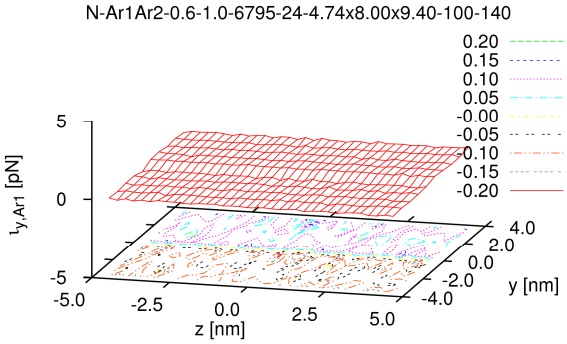

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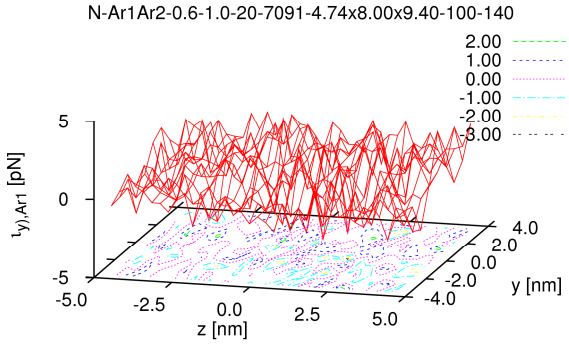
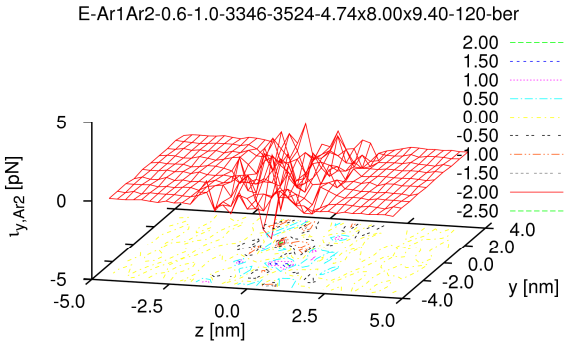
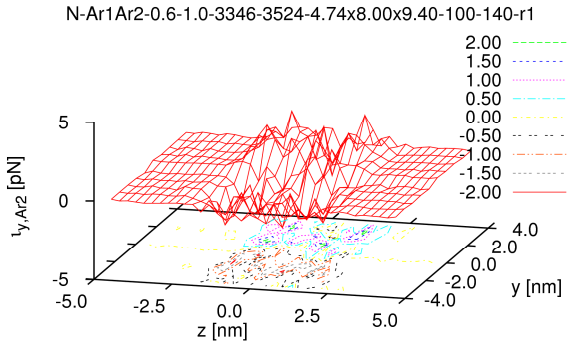


MeansLocYPartForceAr1YZ.eps

i) Interatomic ArA particle *y*-forces



MeansLocYPartForceAr2YZ.eps



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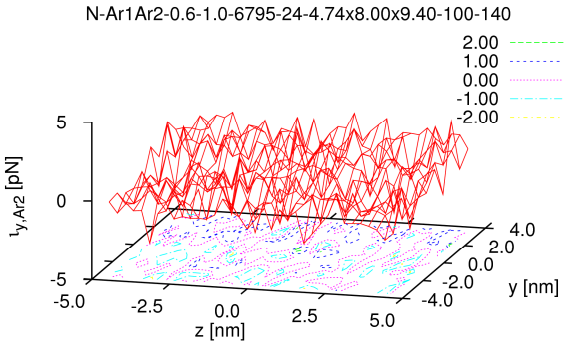

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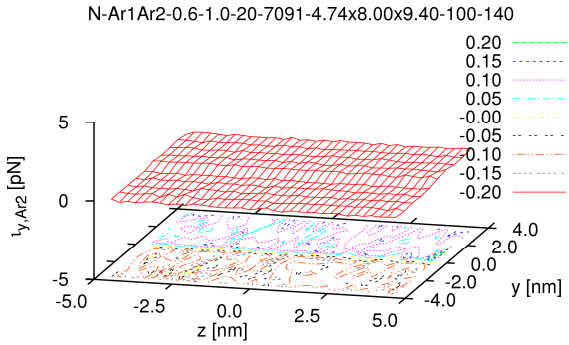
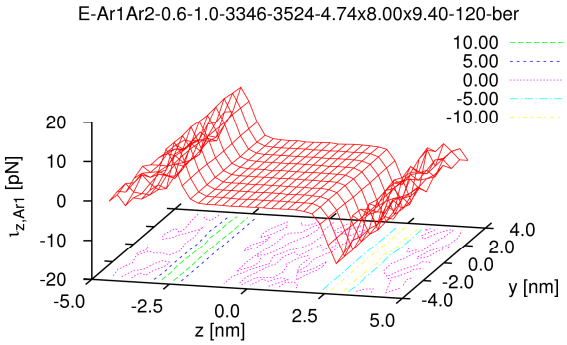


MeansLocYPartForceAr2YZ.eps

j) Interatomic ArB particle *y*-forces



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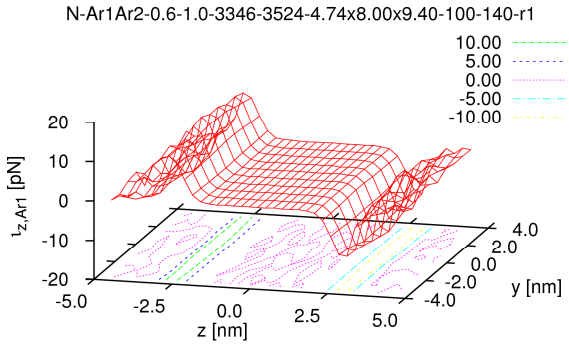

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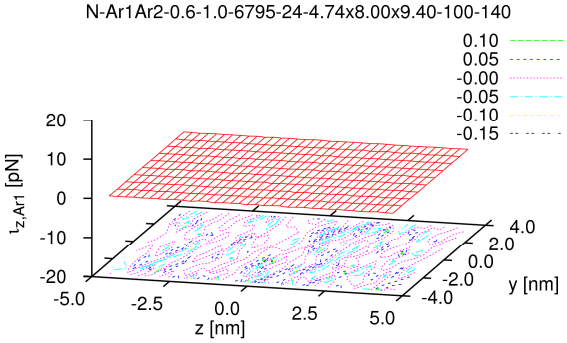

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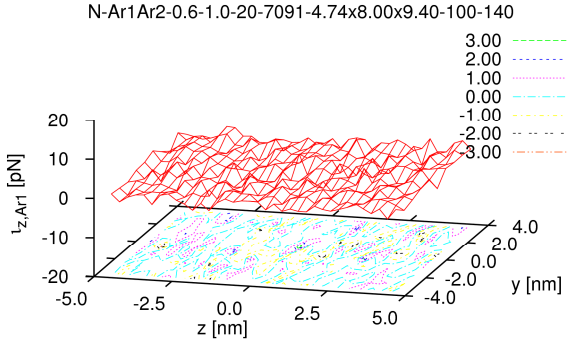

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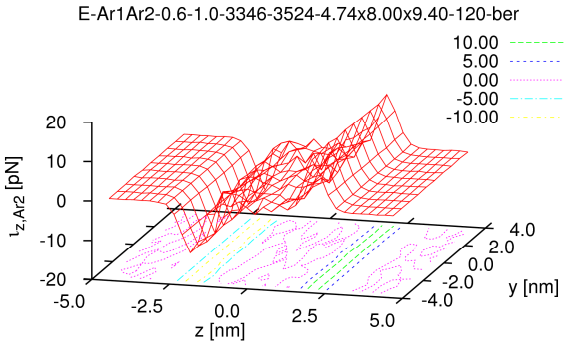
k) Interatomic ArA particle *z*-forces



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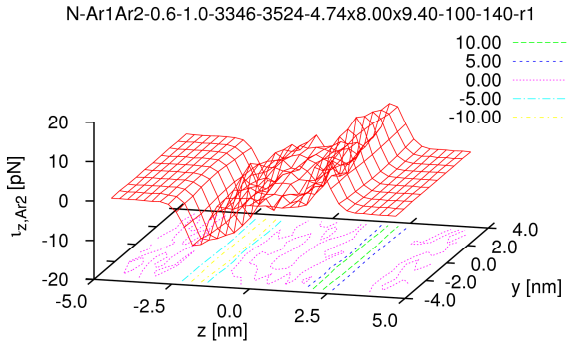
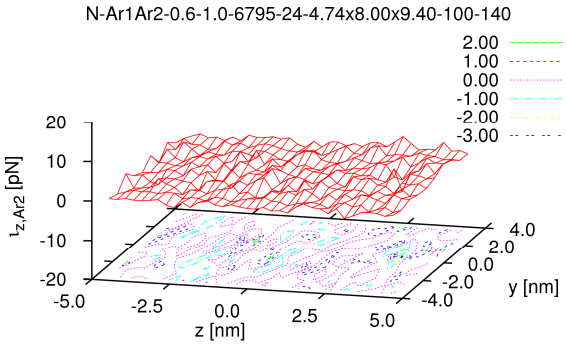


MeansLocZPartForceAr2YZ.eps



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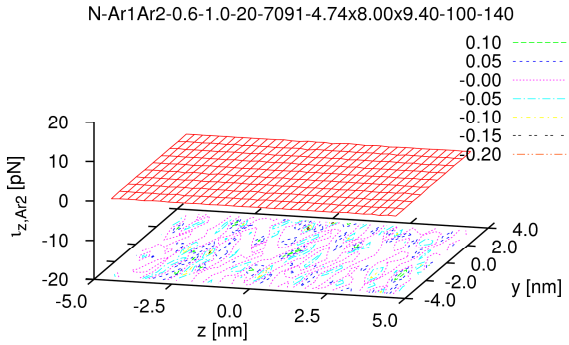
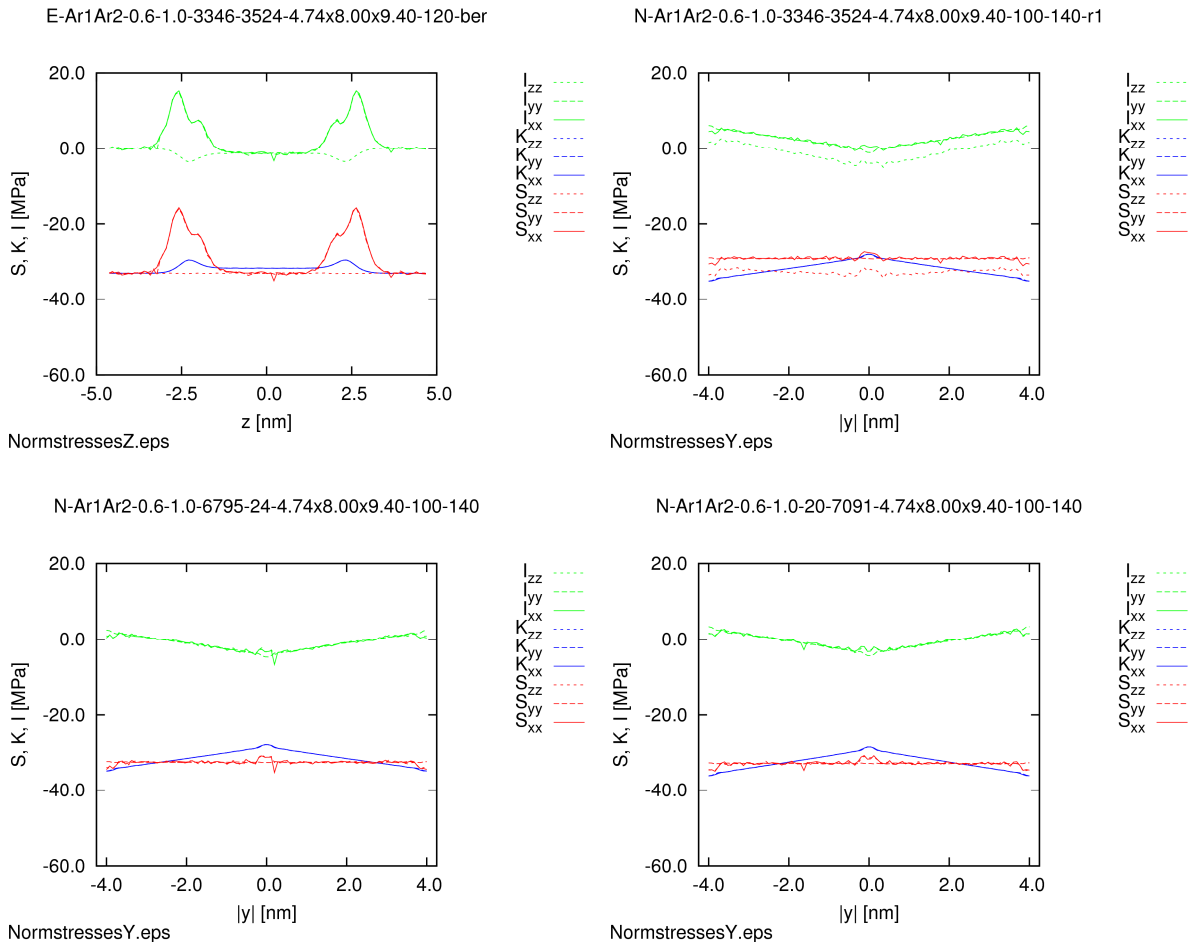


MeansLocZPartForceAr2YZ.eps

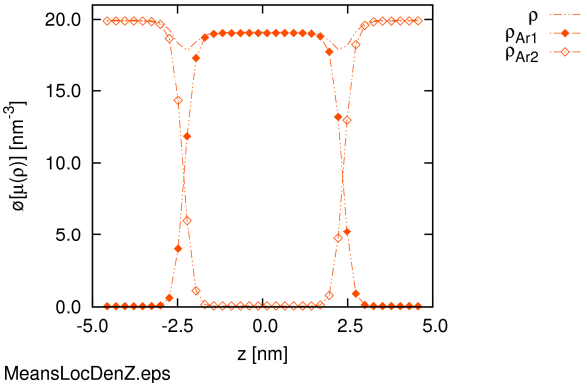
l) Interatomic ArB particle *z*-forces



m) Normal stresses

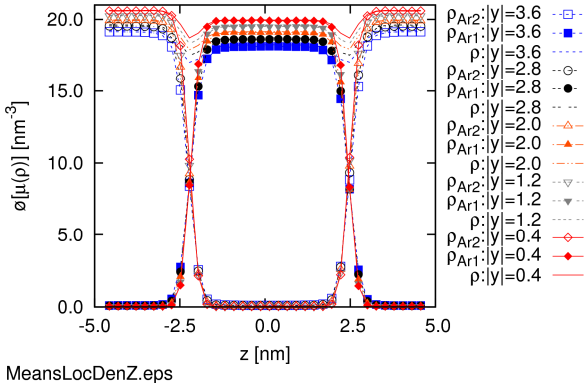
Figure 34 Detailed distributions of the local observables in the first heterophasic nonequilibrium interfacial system and its corresponding ones N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1, E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber, N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140, and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



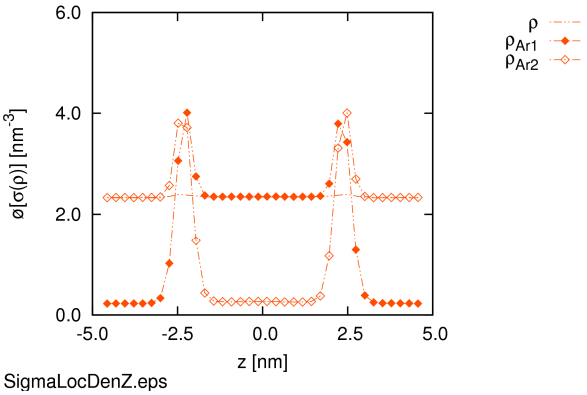
MeansLocDenZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



MeansLocDenZ.eps

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



SigmaLocDenZ.eps

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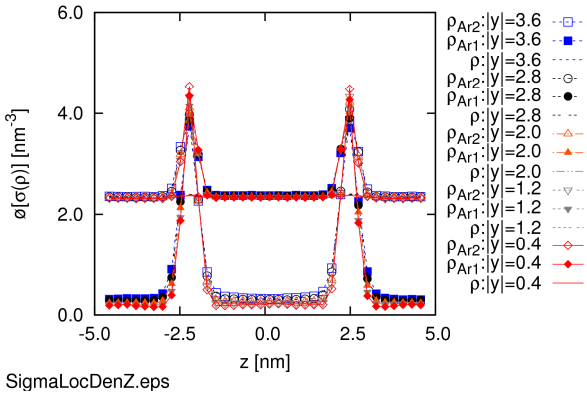
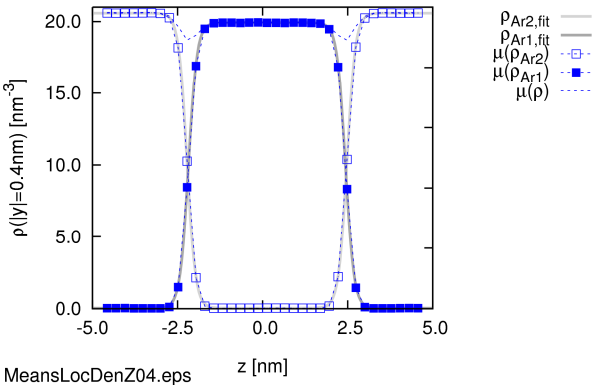


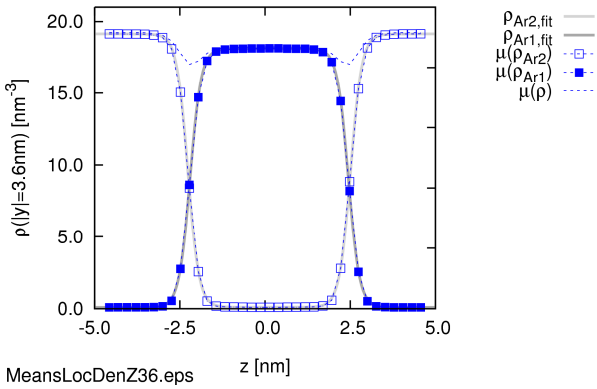
SigmaLocDenZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



MeansLocDenZ04.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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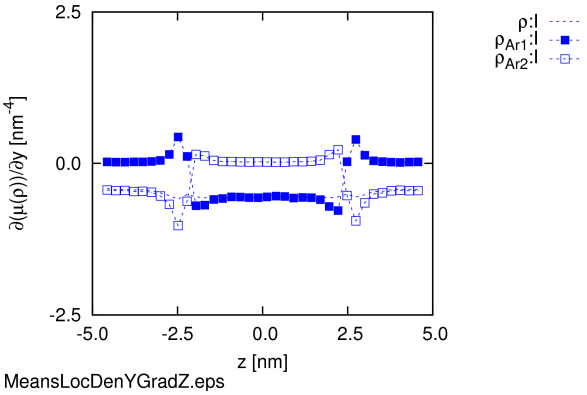
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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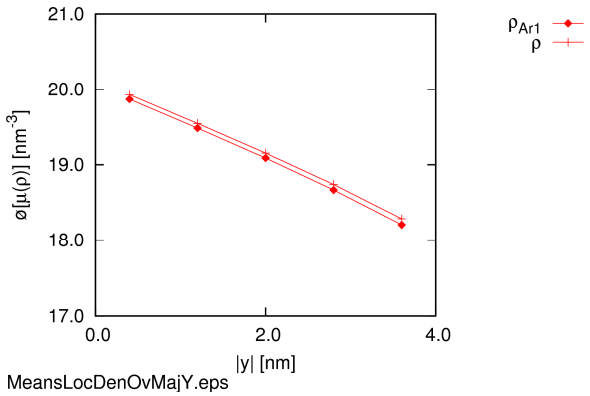
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



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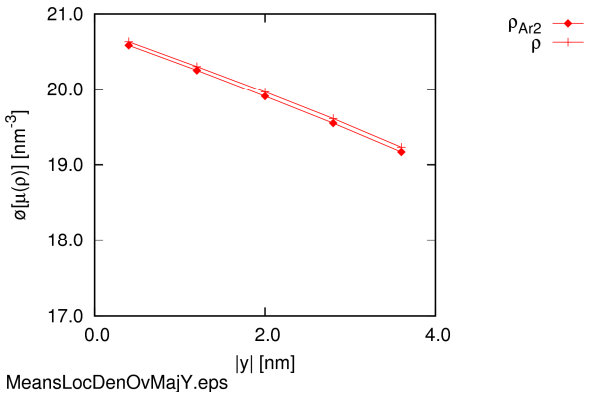
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



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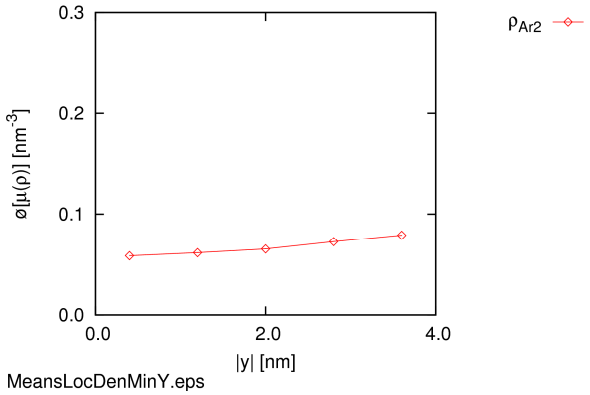

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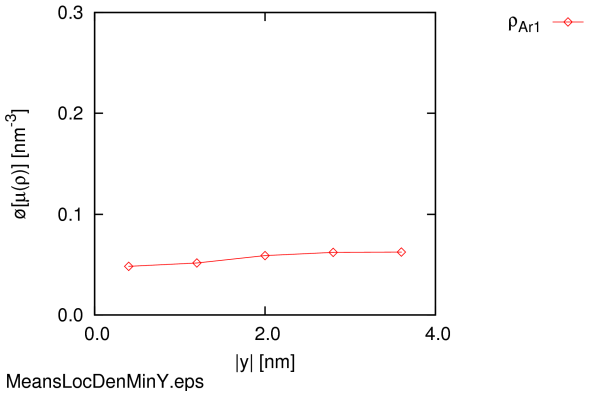

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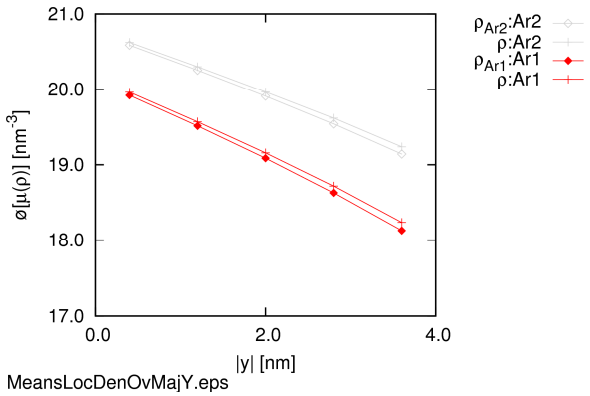
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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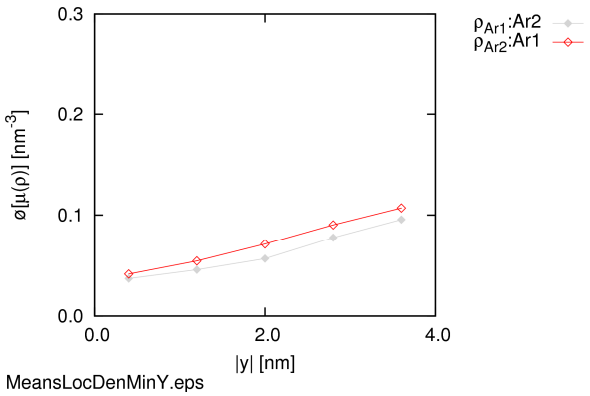

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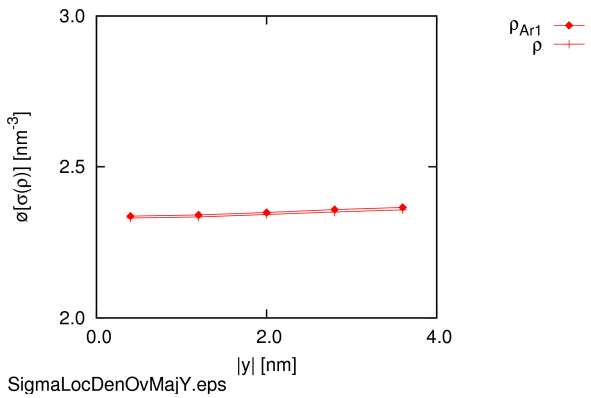

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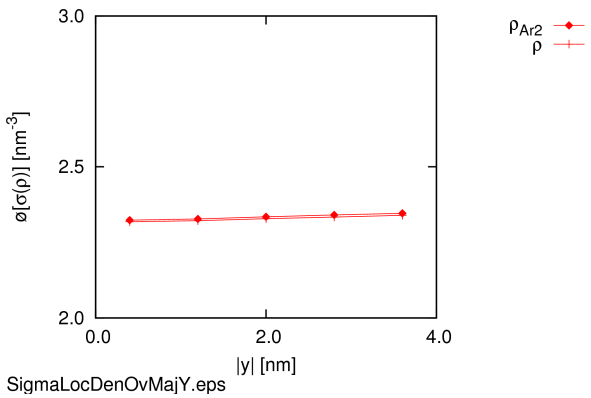

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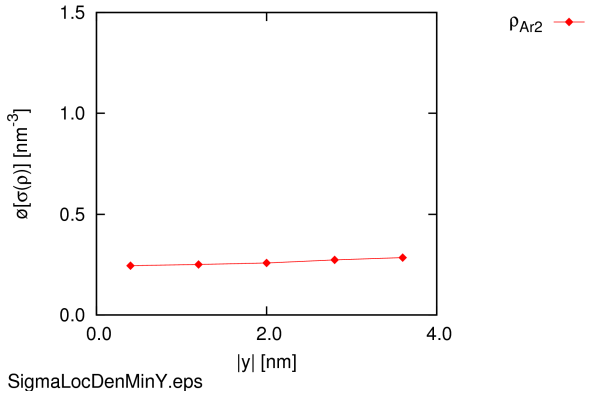
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



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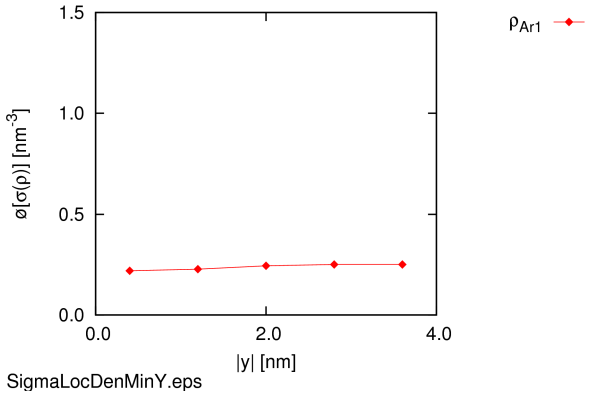
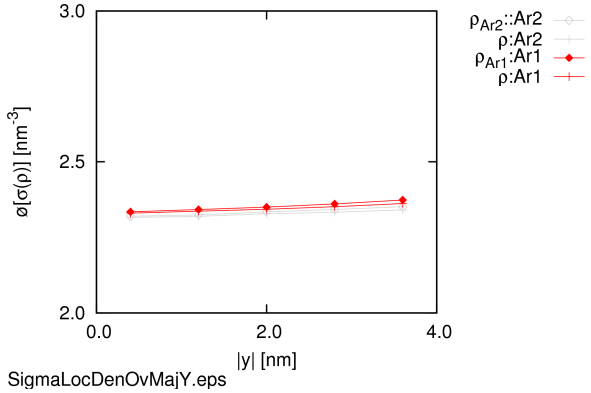
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



SigmaLocDenMinY.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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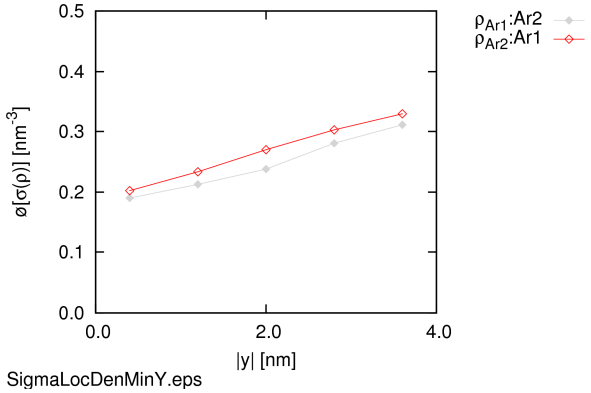
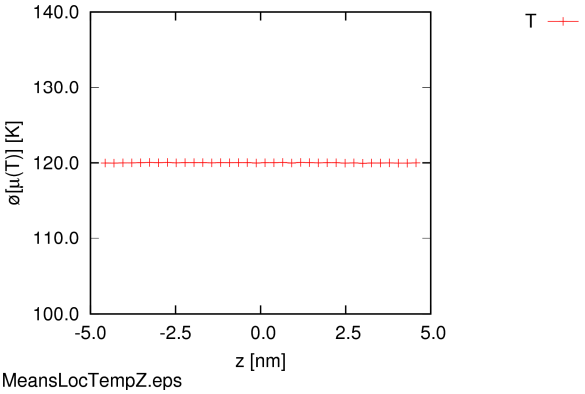
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



SigmaLocDenMinY.eps

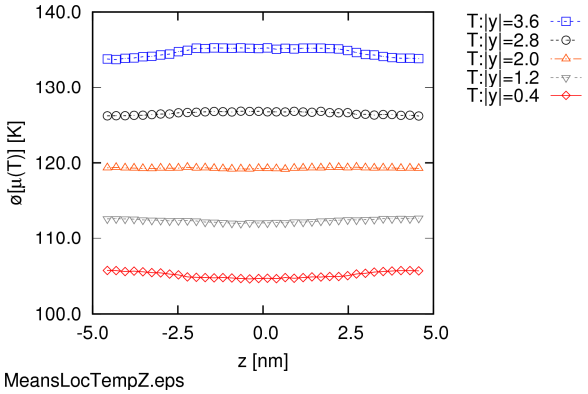
a) Densities

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



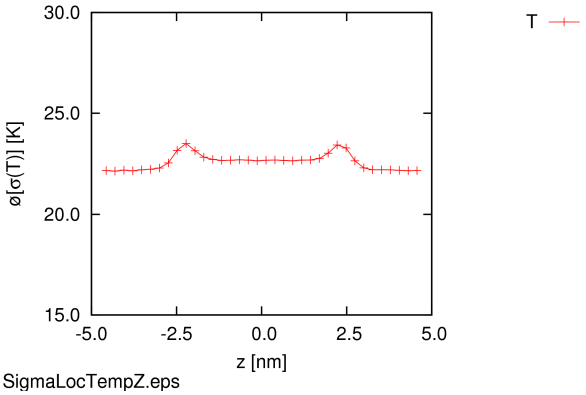
MeansLocTempZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



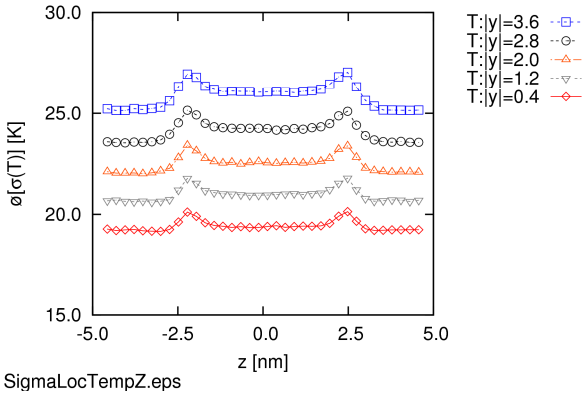
MeansLocTempZ.eps

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



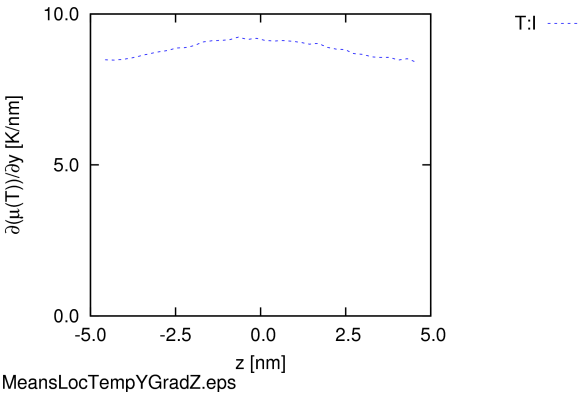
SigmaLocTempZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



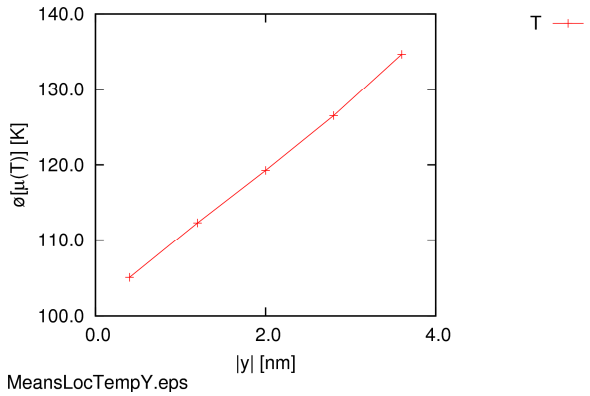
SigmaLocTempZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



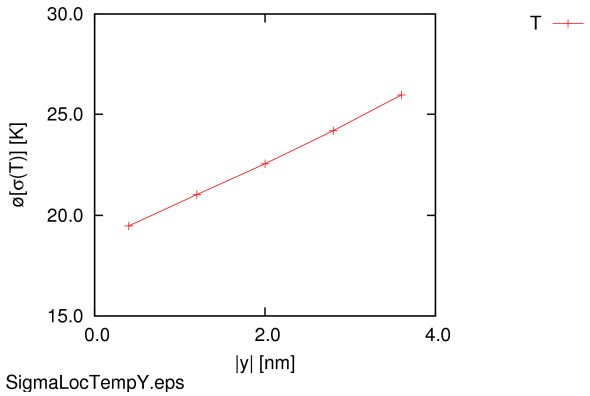
MeansLocTempYGradZ.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



MeansLocTempY.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



SigmaLocTempY.eps

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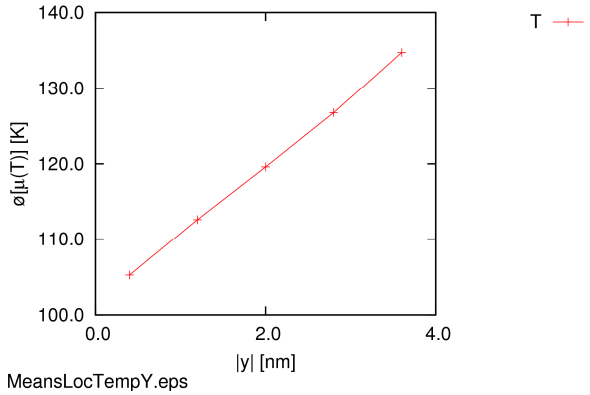

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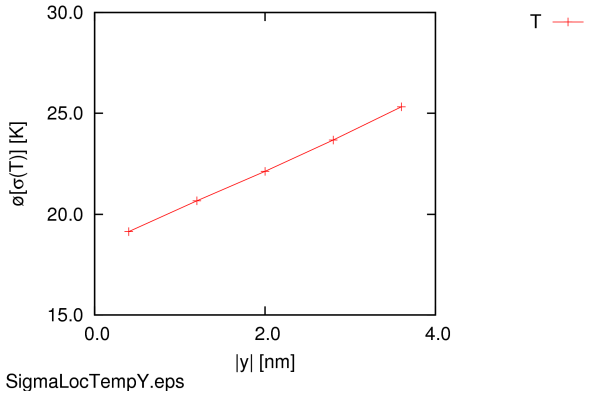
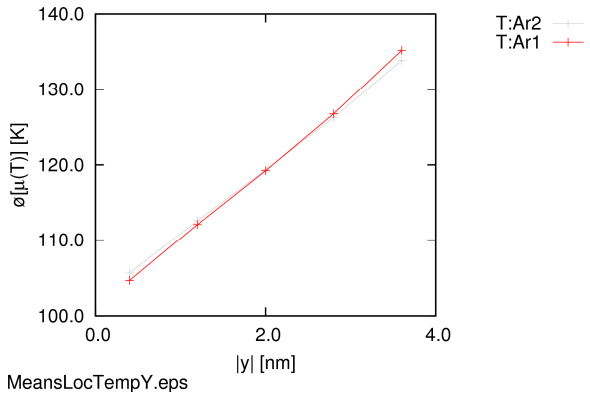
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



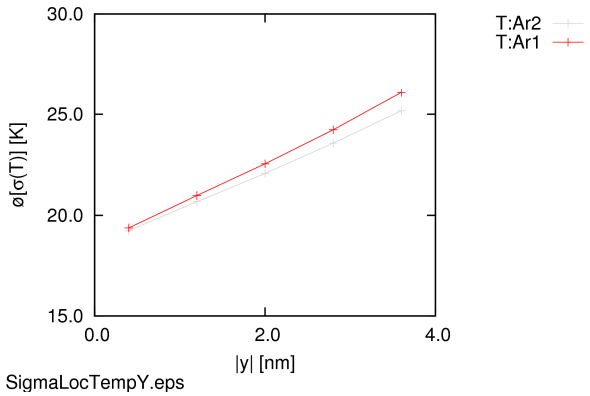
SigmaLocTempY.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



MeansLocTempY.eps

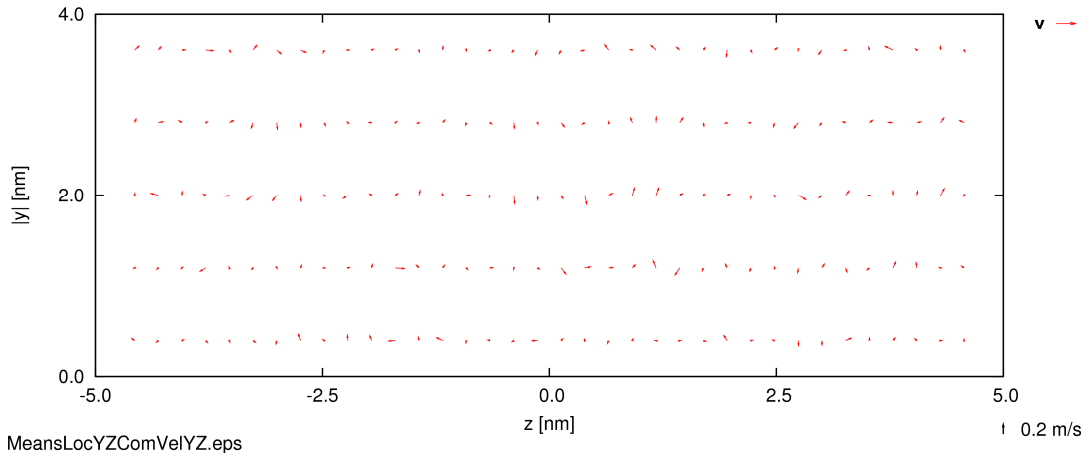
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



SigmaLocTempY.eps

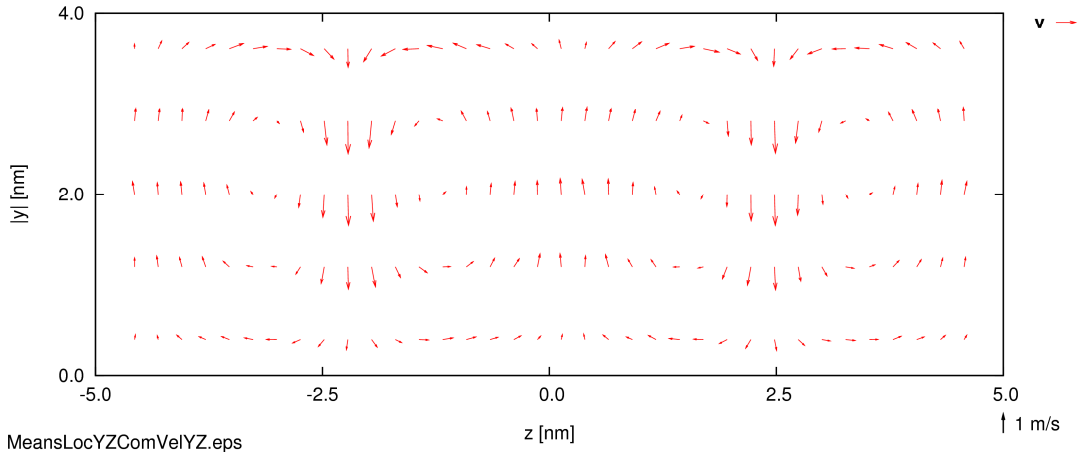
b) Temperatures

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



MeansLocYZComVelYZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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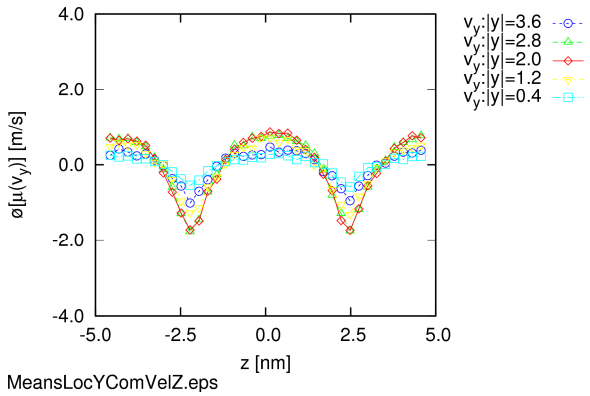
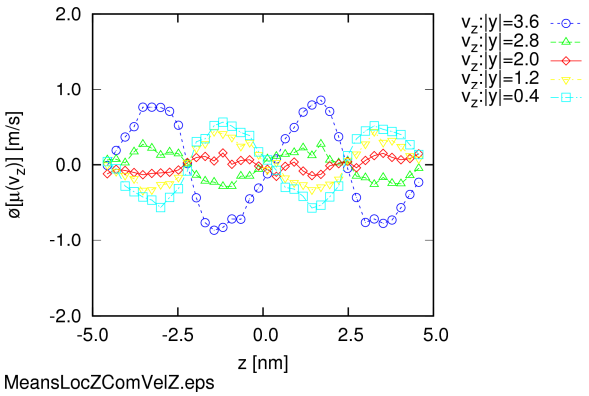


MeansLocYComVelZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



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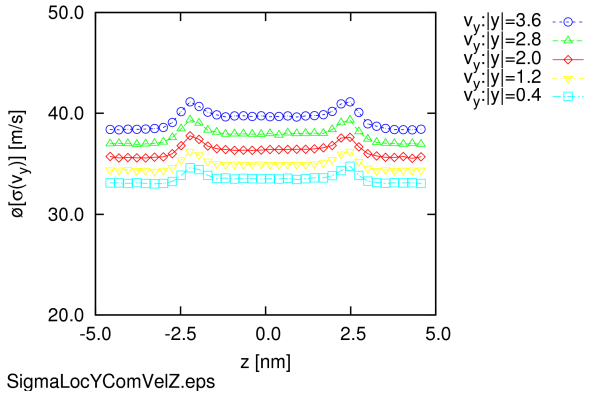
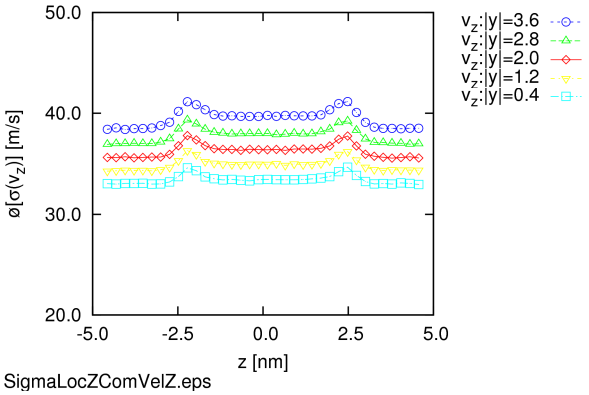
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1

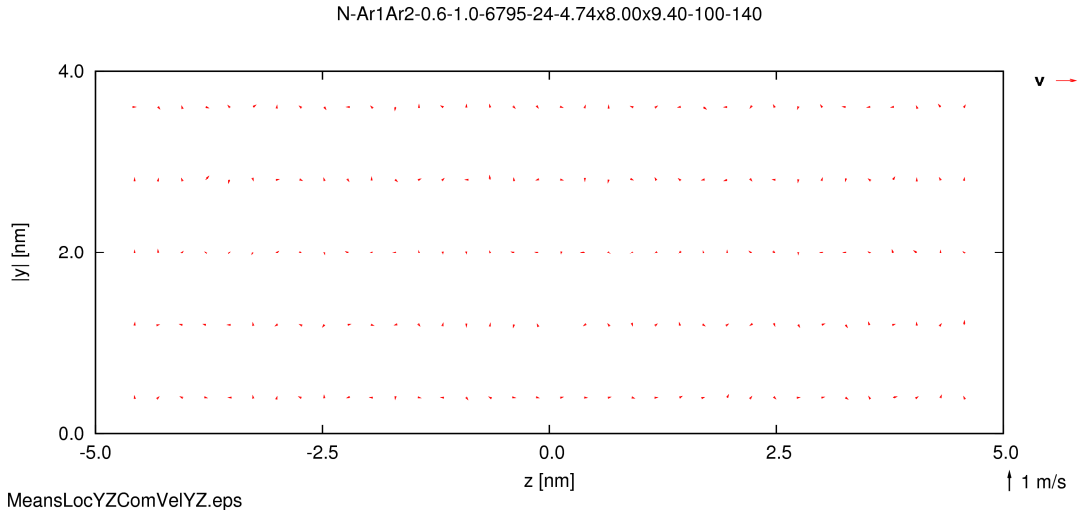


SigmaLocYComVelZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



SigmaLocZComVelZ.eps



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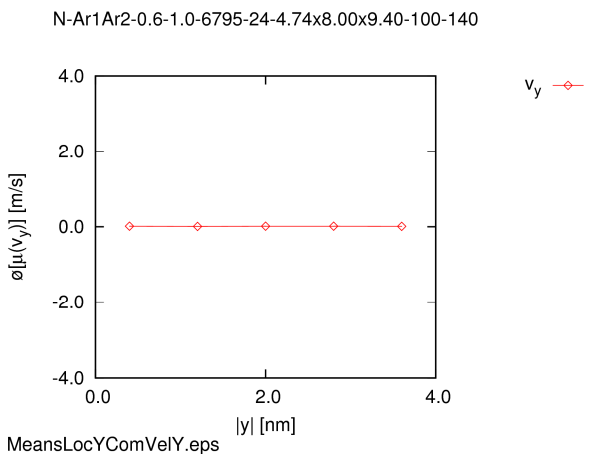

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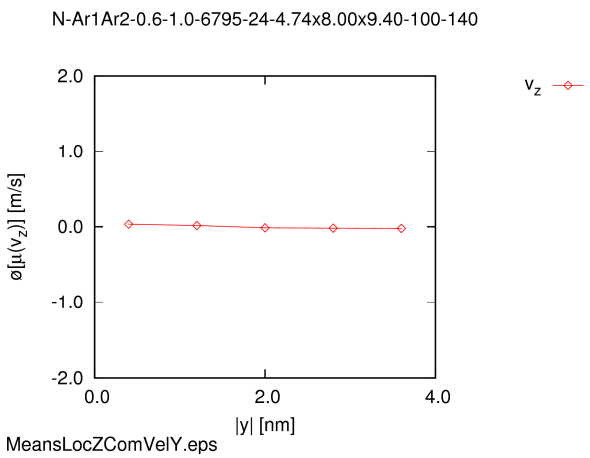

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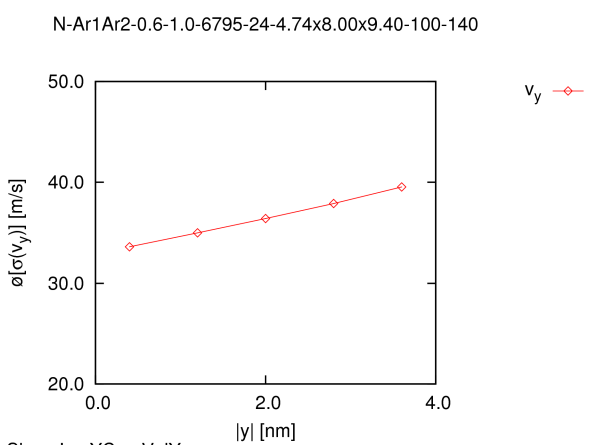

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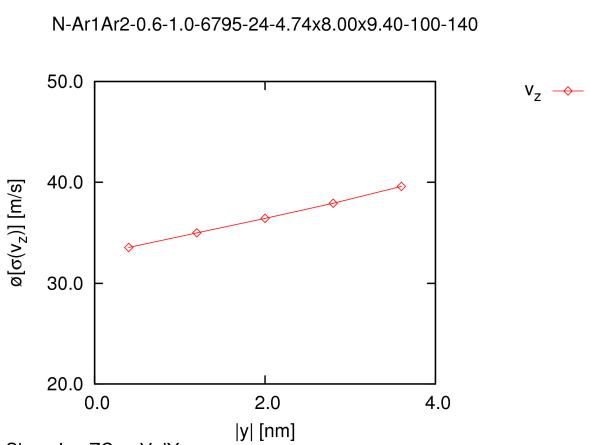

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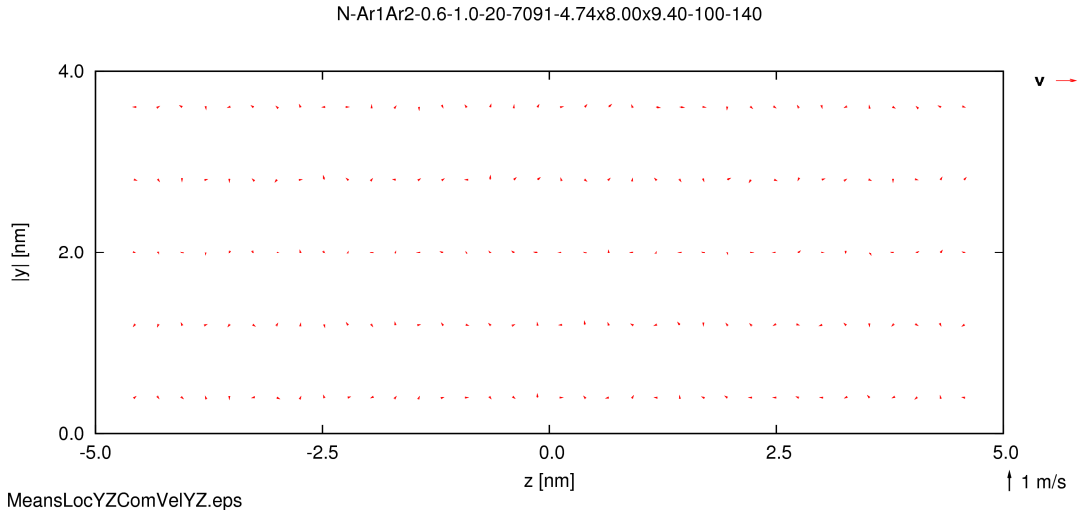

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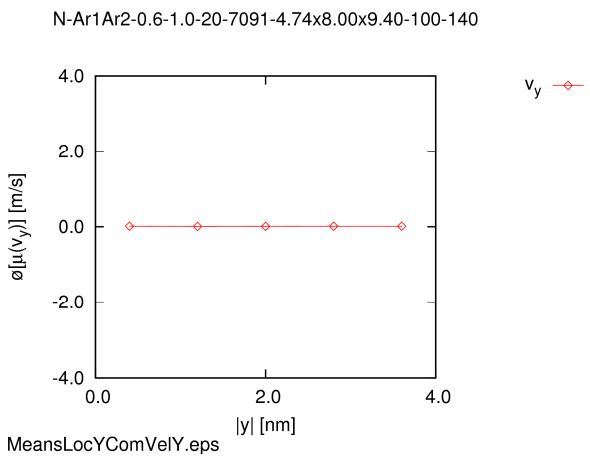

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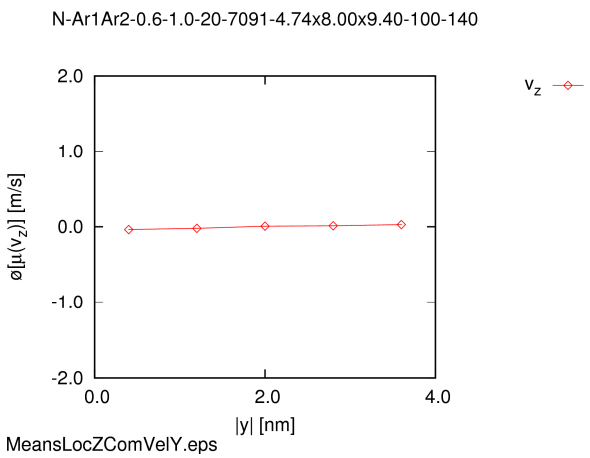

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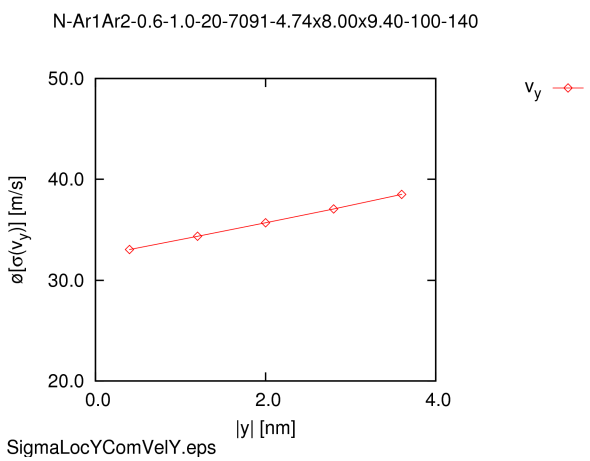
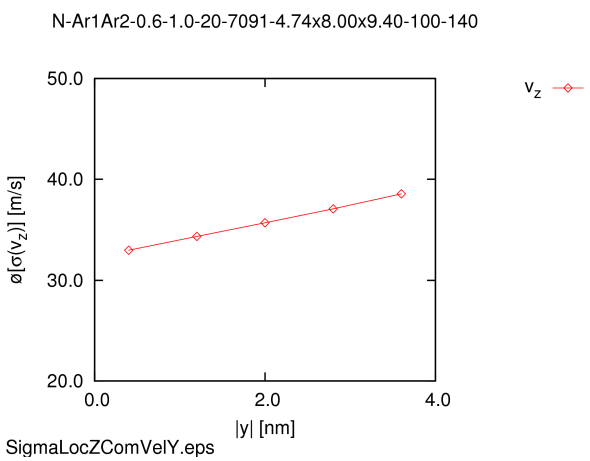


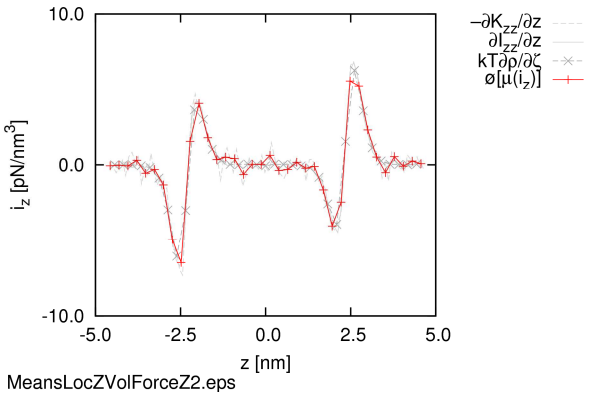
SigmaLocYComVelY.eps



SigmaLocZComVelY.eps

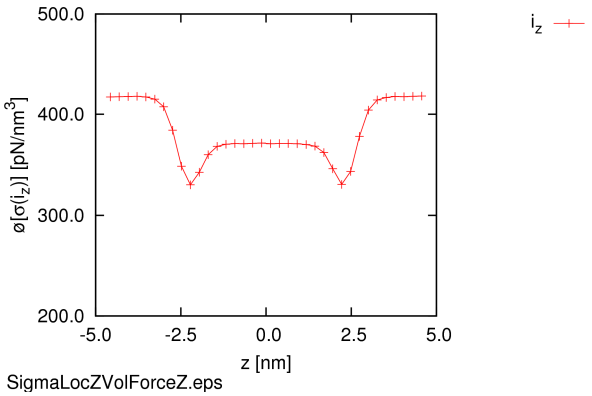
c) Com velocities

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



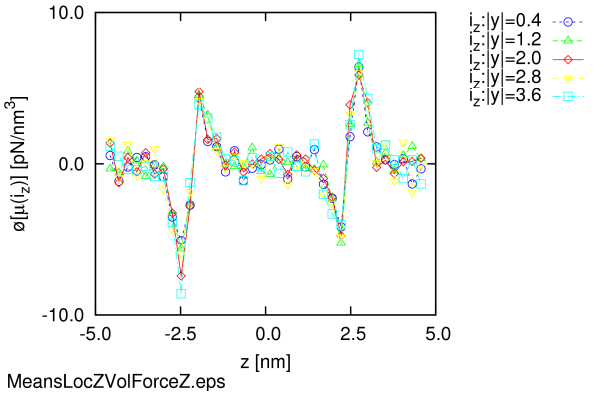
MeansLocZVolForceZ2.eps

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



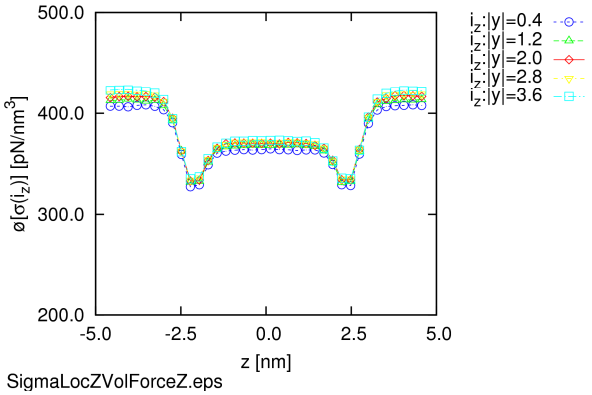
SigmaLocZVolForceZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



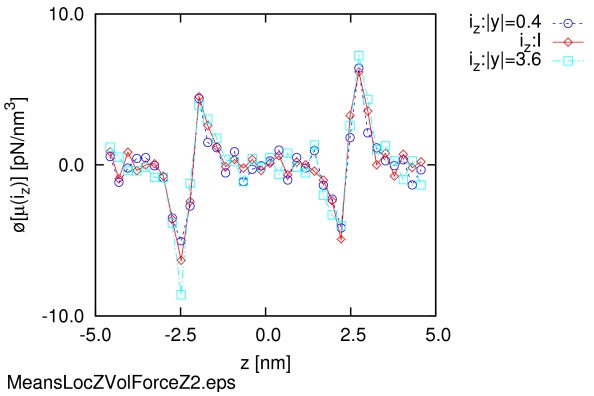
MeansLocZVolForceZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



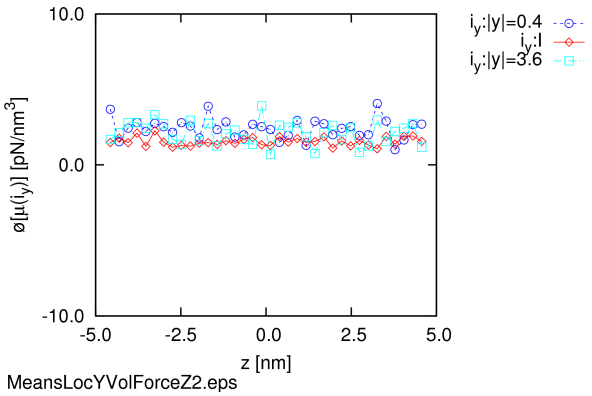
SigmaLocZVolForceZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



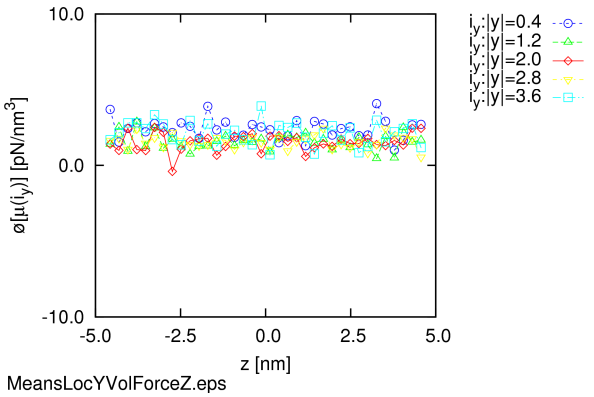
MeansLocZVolForceZ2.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



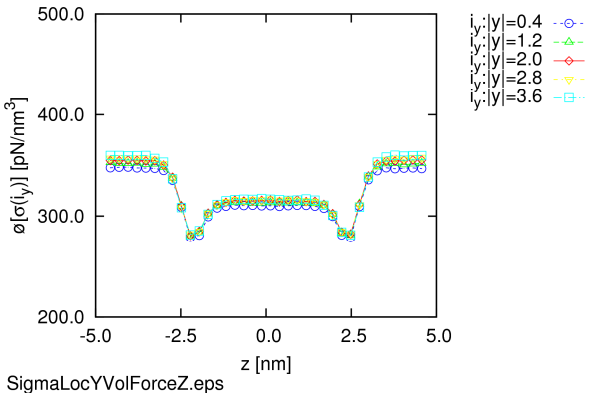
MeansLocYVolForceZ2.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



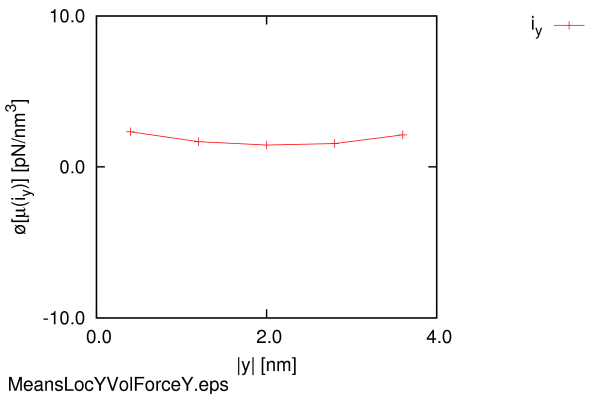
MeansLocYVolForceZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



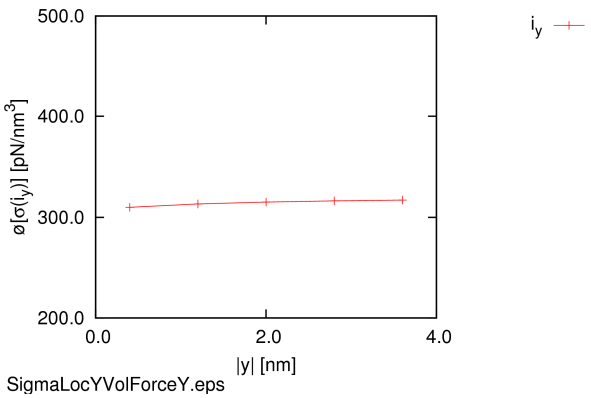
SigmaLocYVolForceZ.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



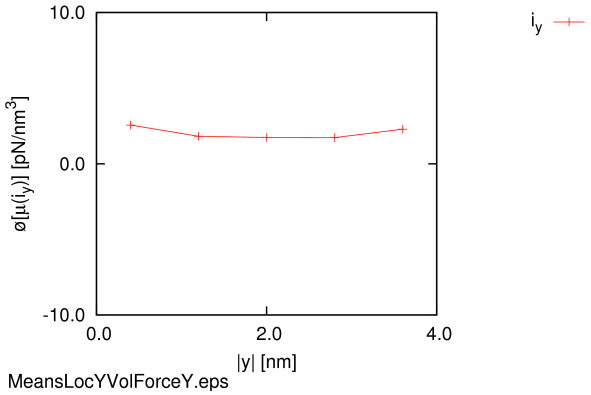
MeansLocYVolForceY.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



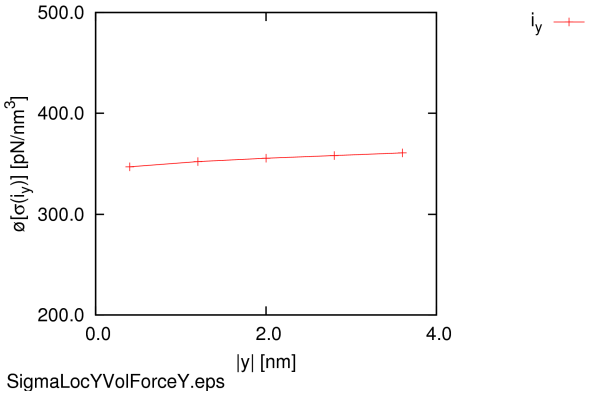
SigmaLocYVolForceY.eps

N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



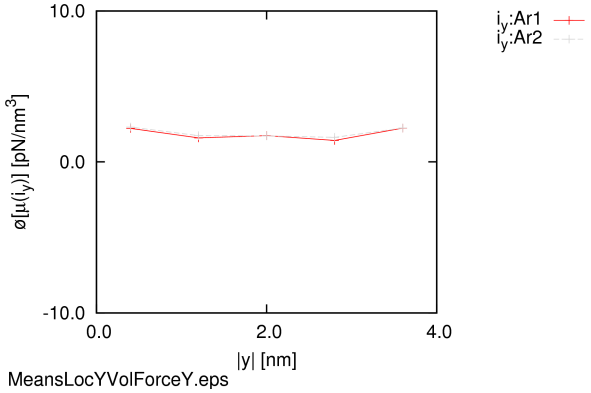
MeansLocYVolForceY.eps

N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



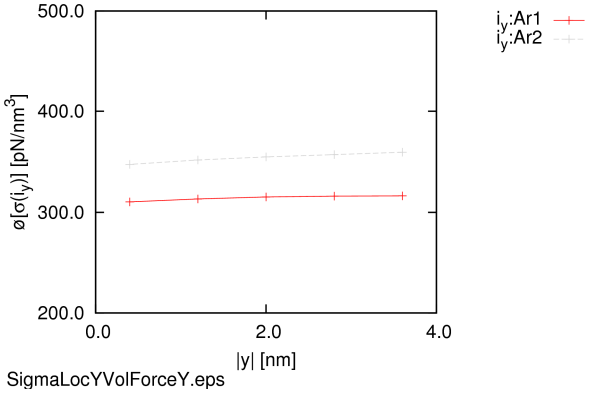
SigmaLocYVolForceY.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1

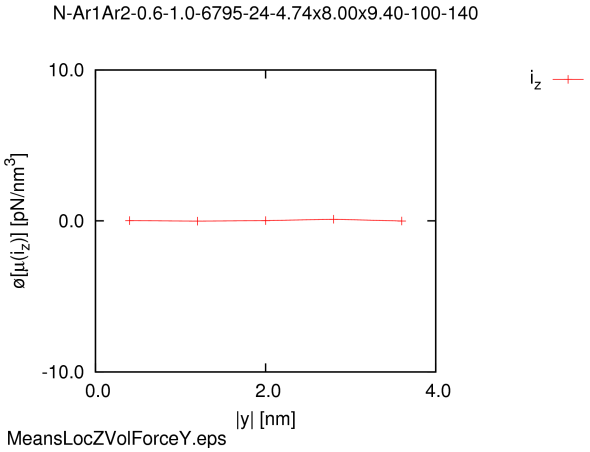


MeansLocYVolForceY.eps

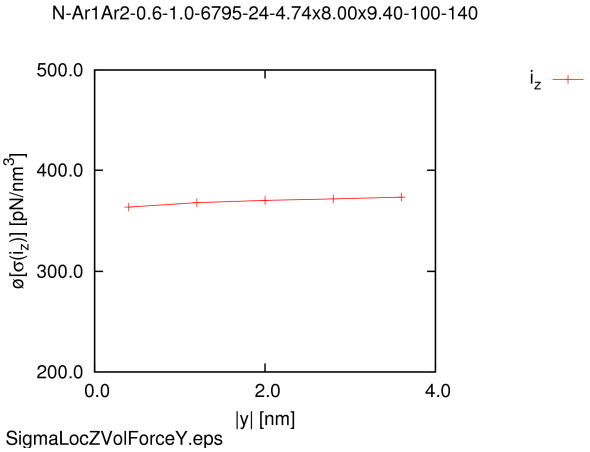
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



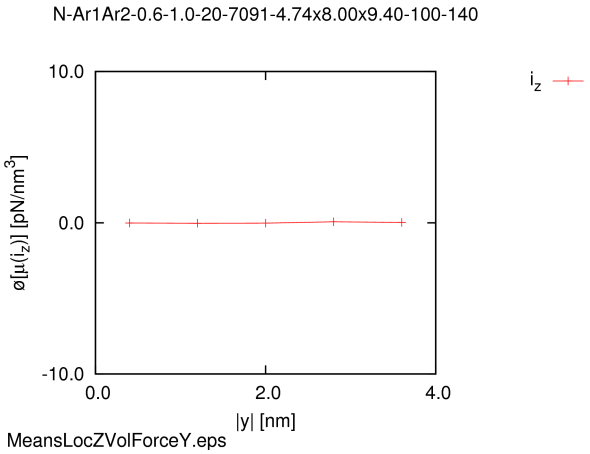
SigmaLocYVolForceY.eps



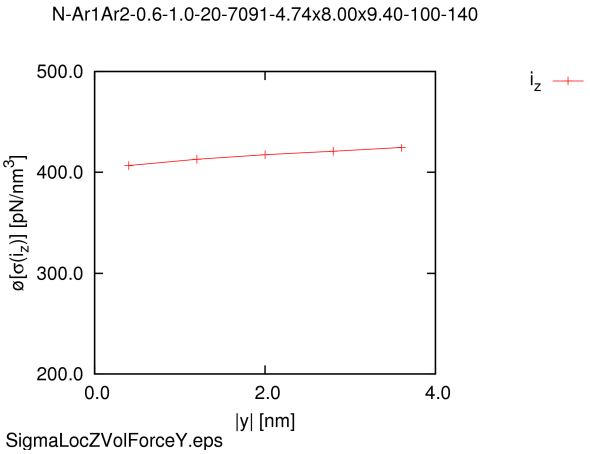
MeansLocZVolForceY.eps



SigmaLocZVolForceY.eps



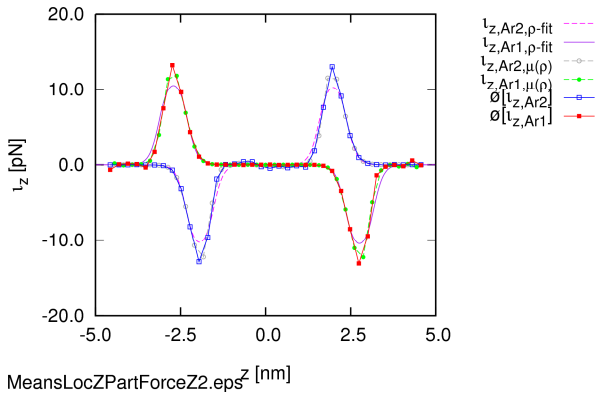
MeansLocZVolForceY.eps



SigmaLocZVolForceY.eps

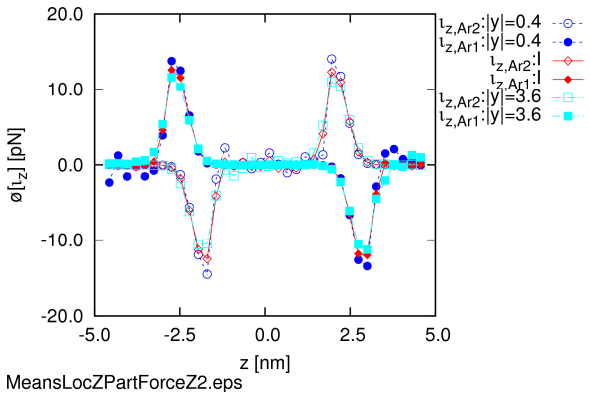
d) Volume forces

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber



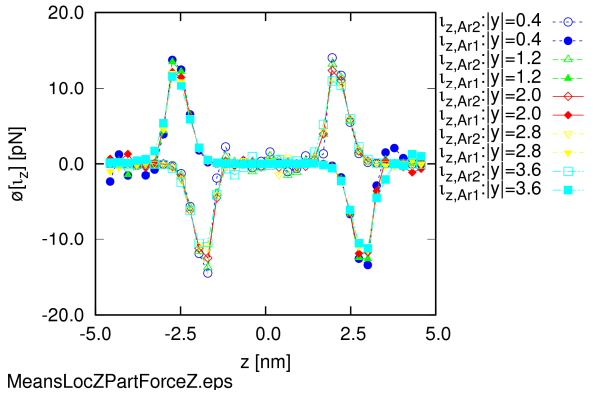
MeansLocZPartForceZ2.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



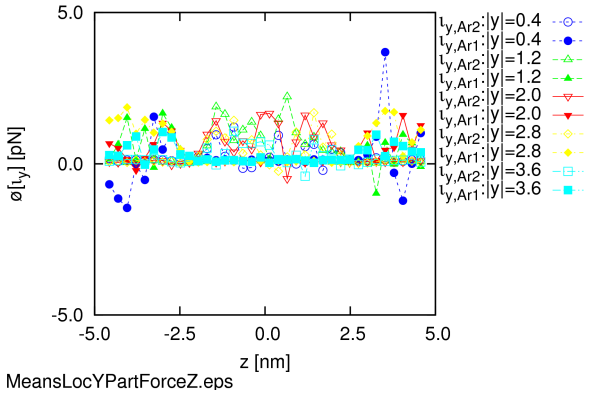
MeansLocZPartForceZ2.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



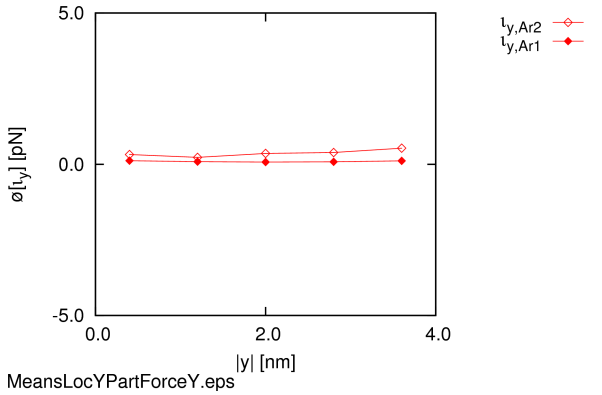
MeansLocZPartForceZ.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



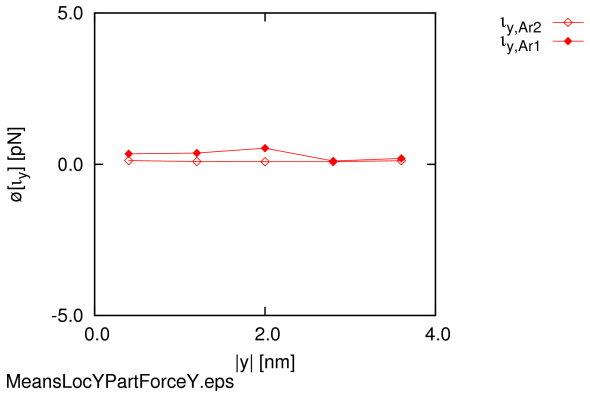
MeansLocYPartForceZ.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140



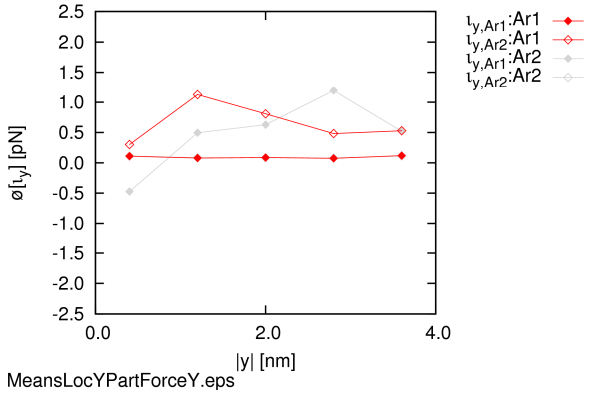
MeansLocYPartForceY.eps

N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140



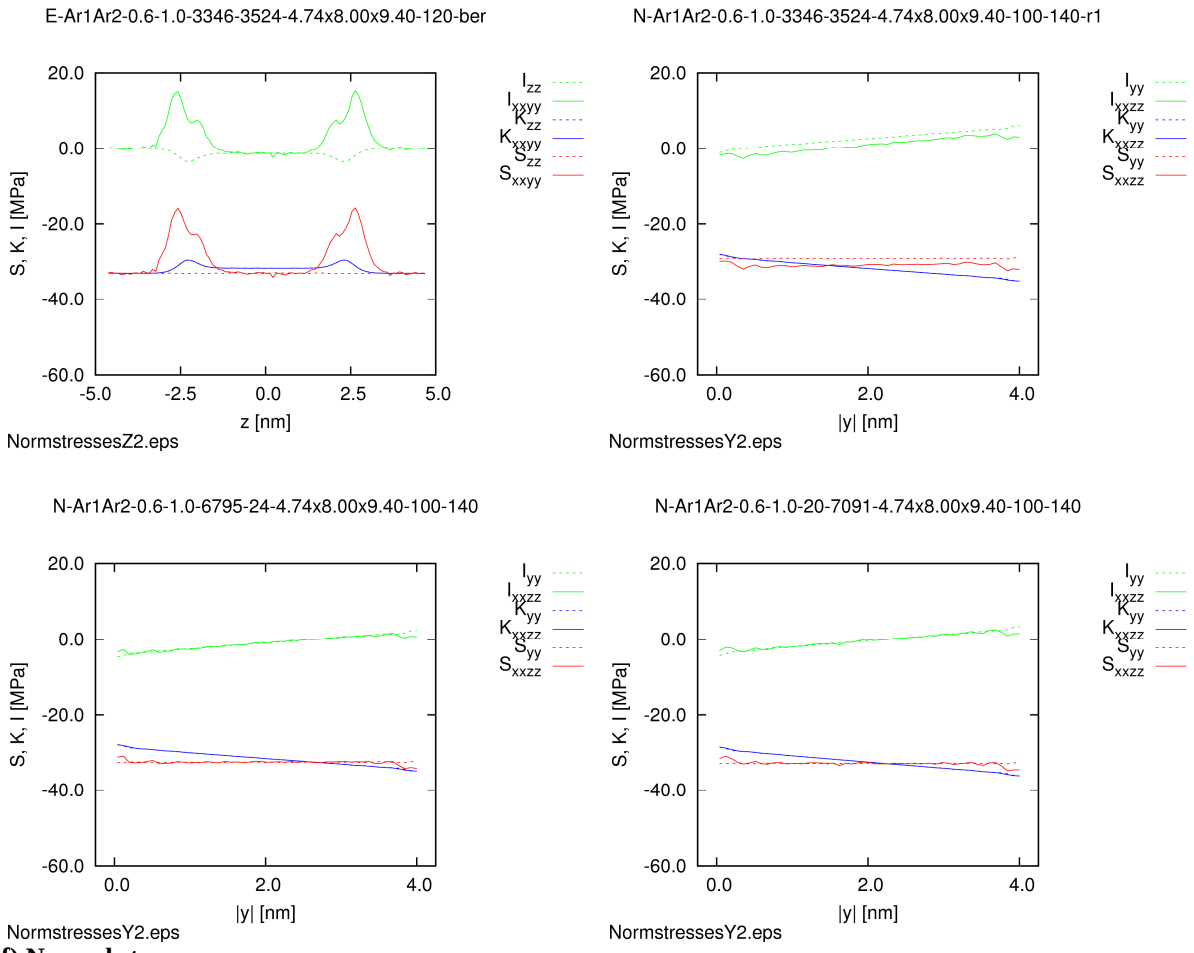
MeansLocYPartForceY.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



MeansLocYPartForceY.eps

e) Particle forces



f) Normal stresses

Figure 35 **Spatially averaged distributions of the local observables in the first heterophasic nonequilibrium interfacial system and its corresponding ones N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1, E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber, N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140, and N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 We plot additionally the fitted hyperbolic tangent density *z*-profiles in the interfacial systems.**

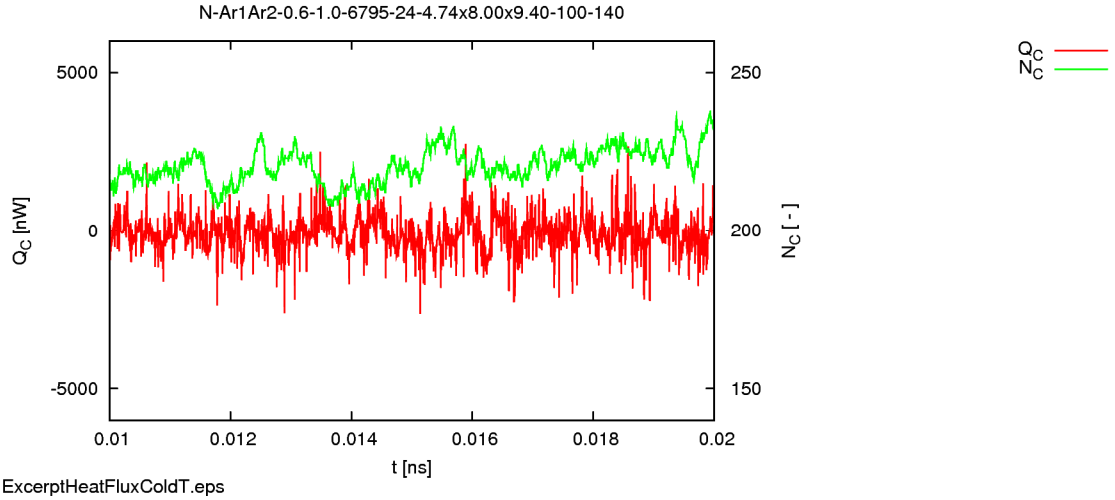
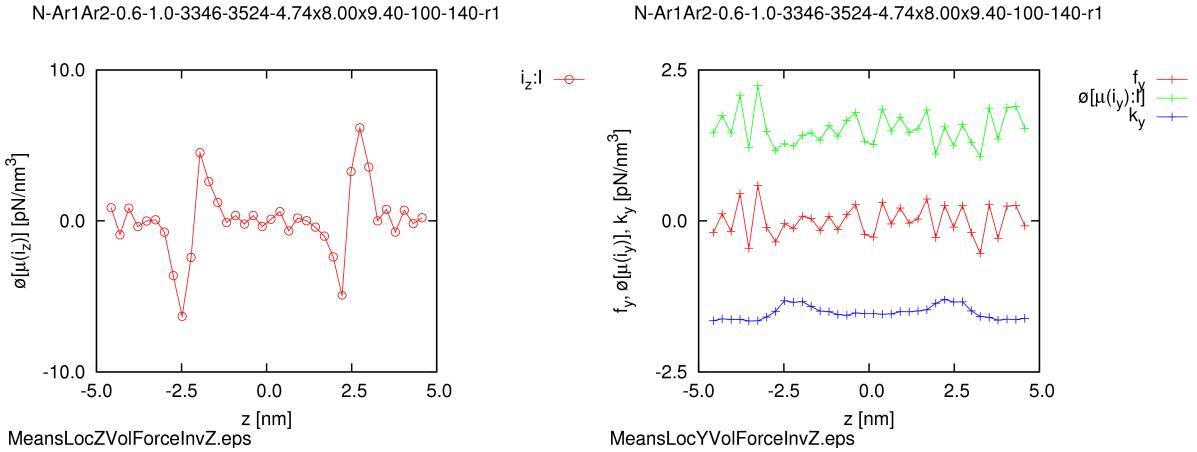
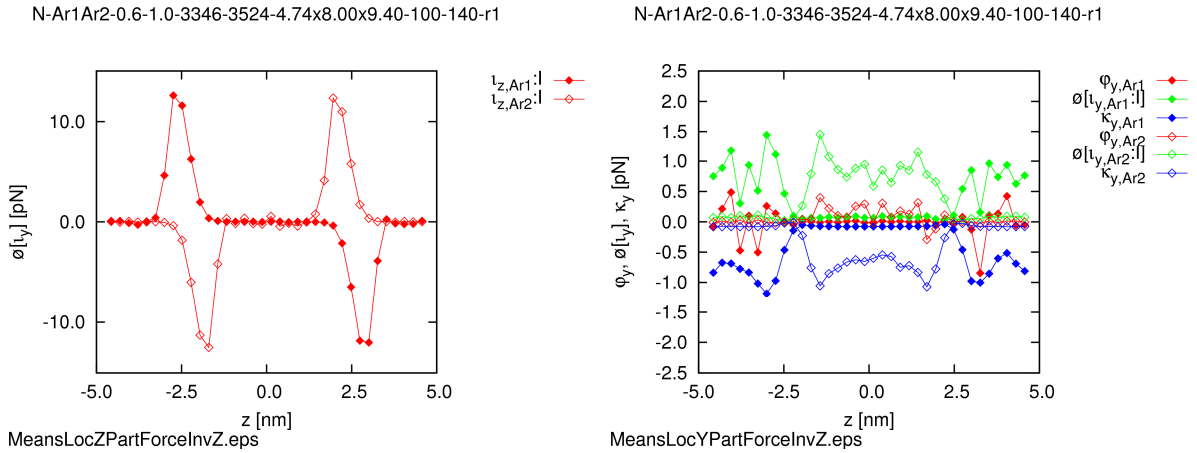


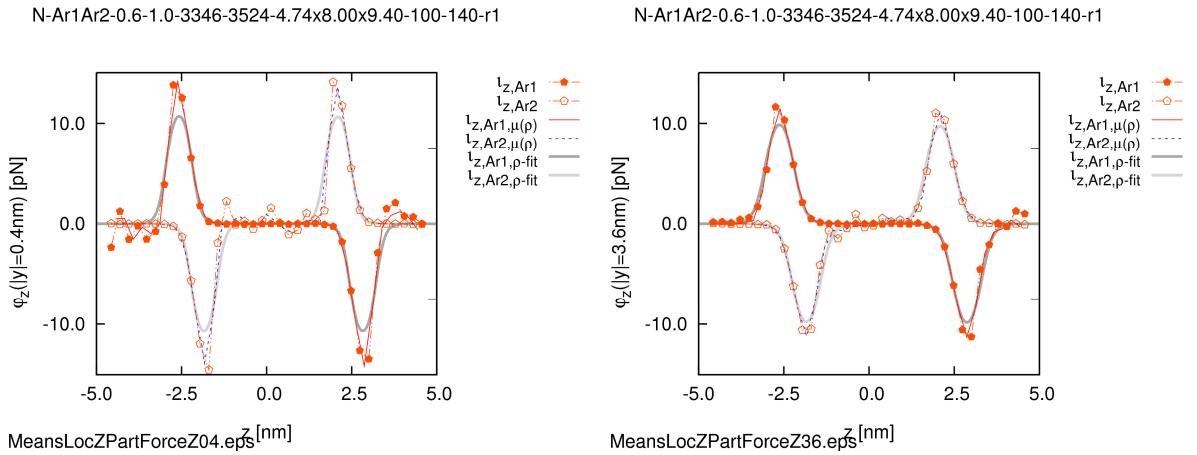
Figure 36 **Excerpt of the instantaneous heat fluxes coupled into or out of the hot thermostated region and the numbers of the particles residing in it in N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140**



a) I-averages of the interatomic and kinetic volume *y*-forces as well as their sum as function of z

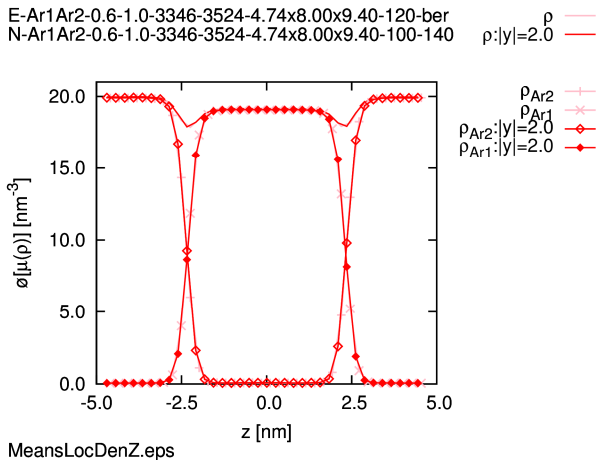


b) I-averages of the interatomic and kinetic particle *y*-forces as well as their sum as function of z

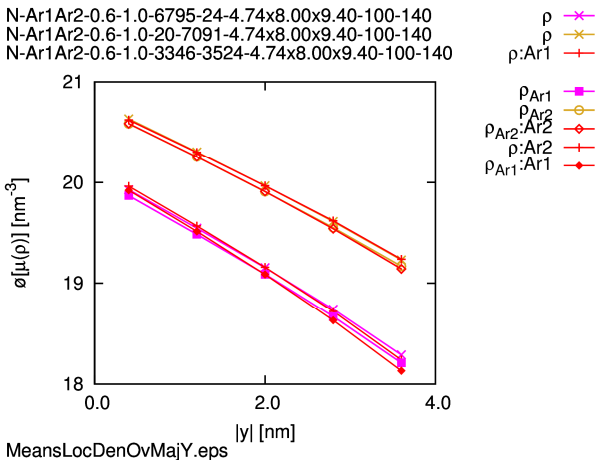


c) Particle *z*-forces obtained in the simulation compared to those computed from the partial density profiles by using the BGY equation using the absolute *y*-coordinates of |y|=0.4nm and of |y|=3.6nm as an example

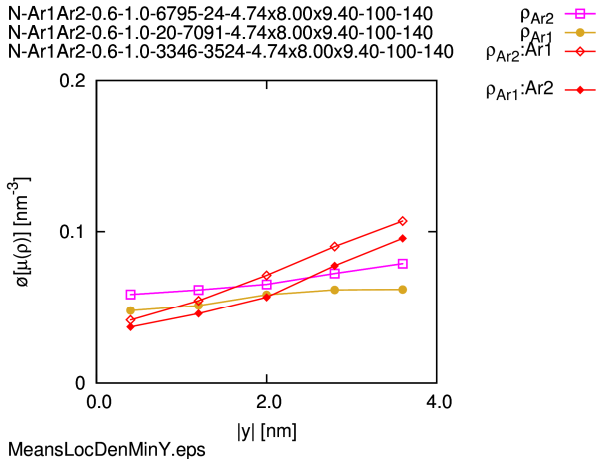
Figure 37 Consistency of the local observables in the first heterophasic nonequilibrium interfacial system N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1



MeansLocDenZ.eps

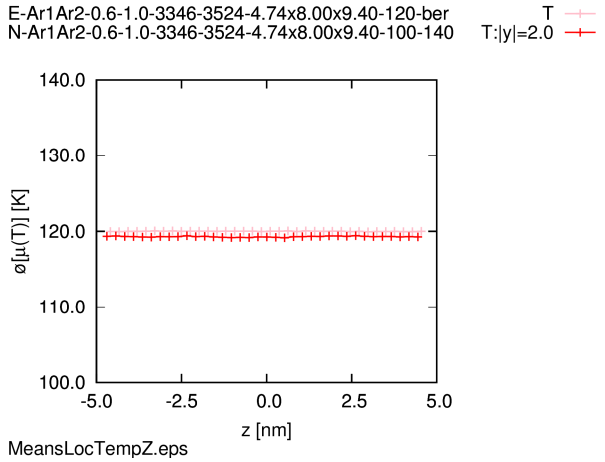


MeansLocDenOvMajY.eps

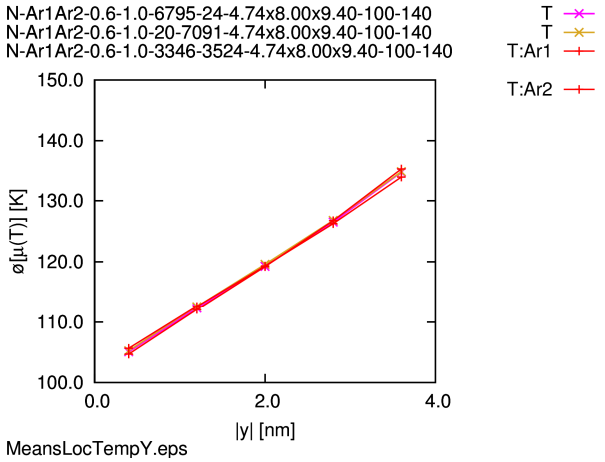


MeansLocDenMinY.eps

a) Densities

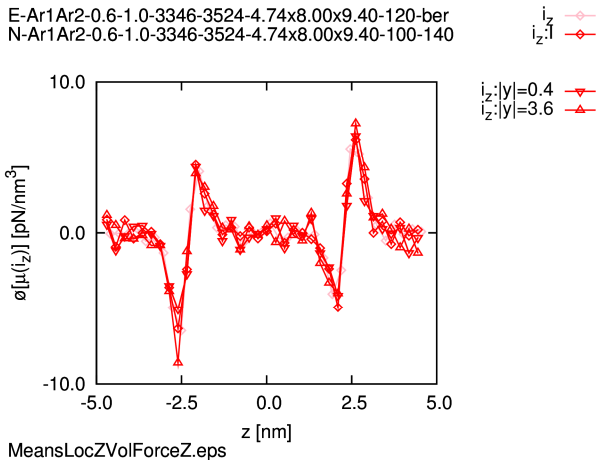


MeansLocTempZ.eps

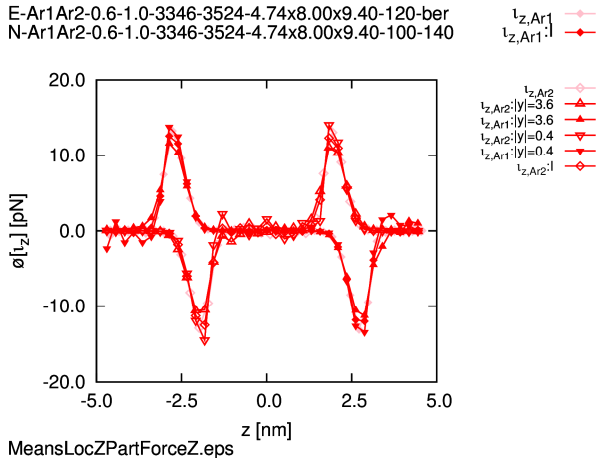


MeansLocTempY.eps

b) Temperatures

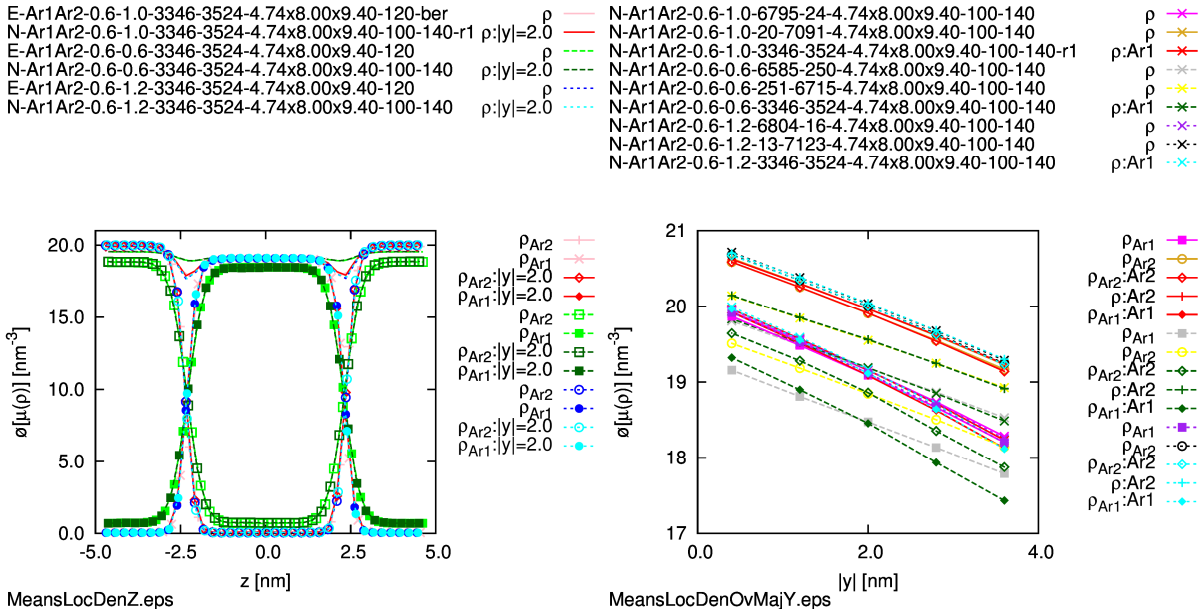


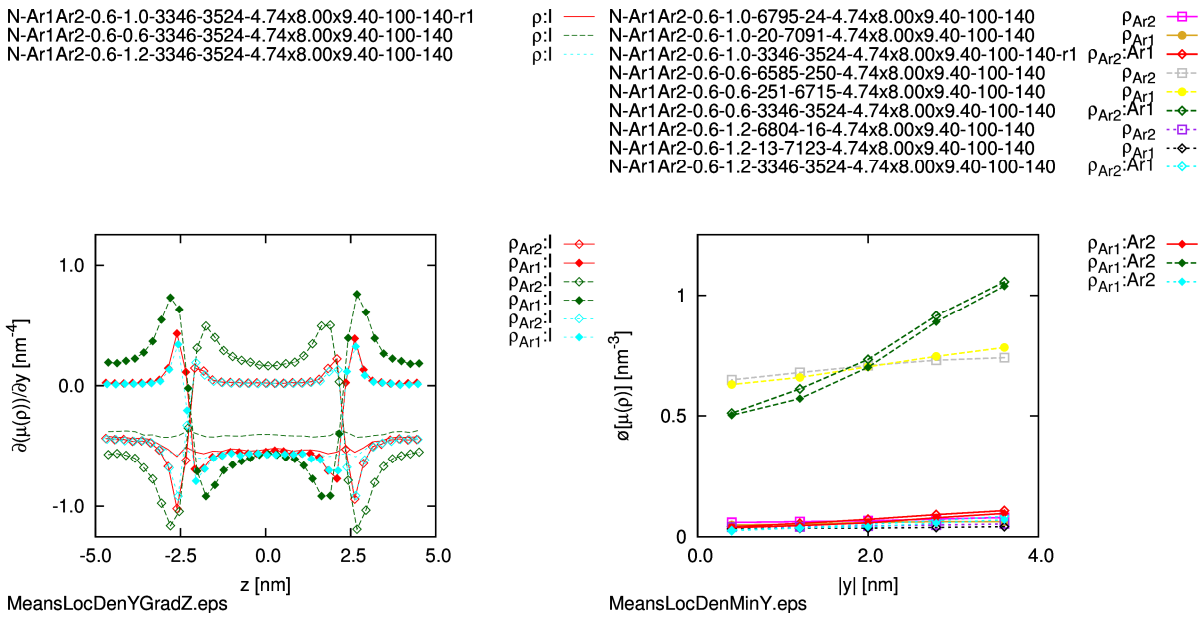
c) Volume forces

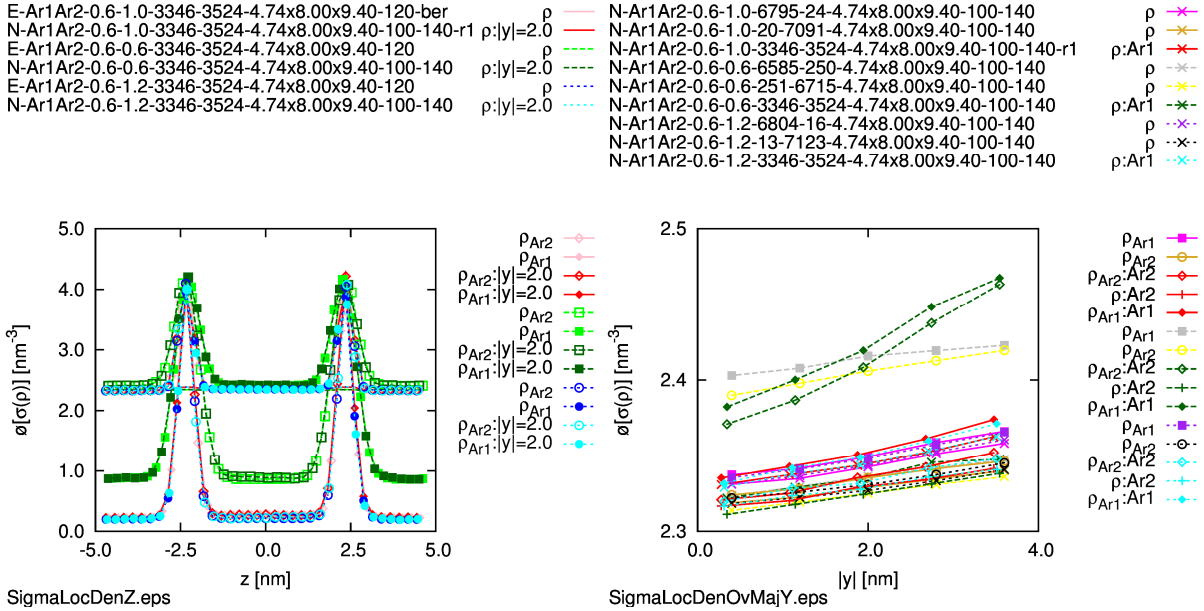


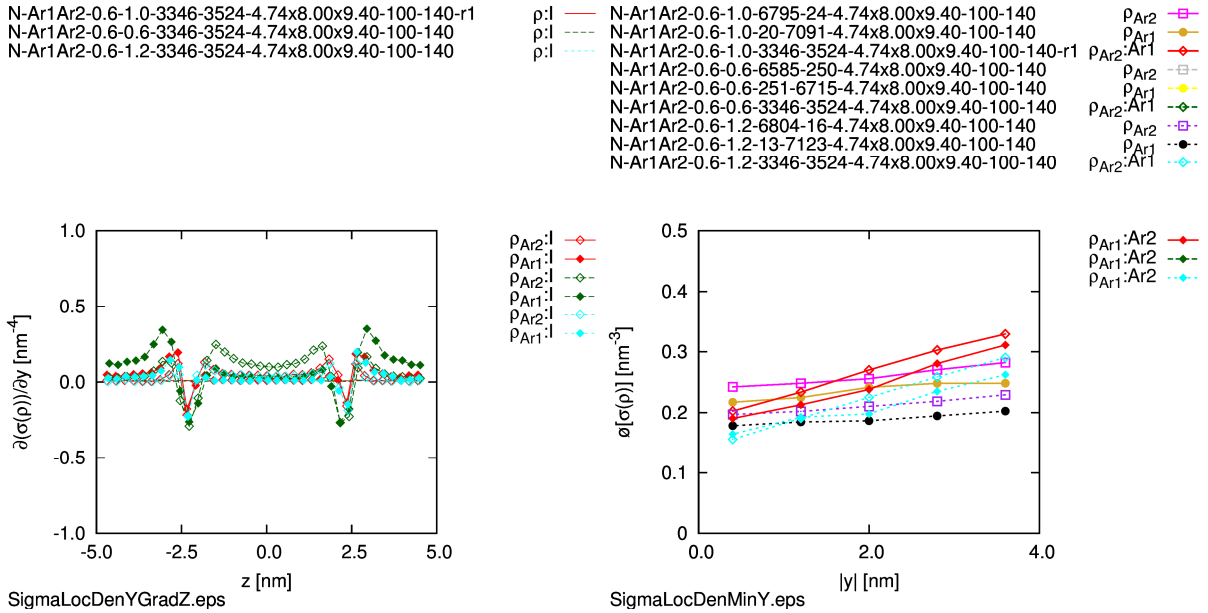
d) Particle forces

Figure 38 Comparison of the summarised local observables in the corresponding first heterophasic interfacial systems E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120 and N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140


E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:Ar1
N-Ar1Ar2-0.6-0.6-6585-250-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-0.6-251-6715-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
N-Ar1Ar2-0.6-1.2-6804-16-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.2-13-7123-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
ø[μ(ρ)] [nm-3]
z [nm]
MeansLocDenZ.eps
ø[μ(ρ)] [nm-3]
|y| [nm]
MeansLocDenOvMajY.eps

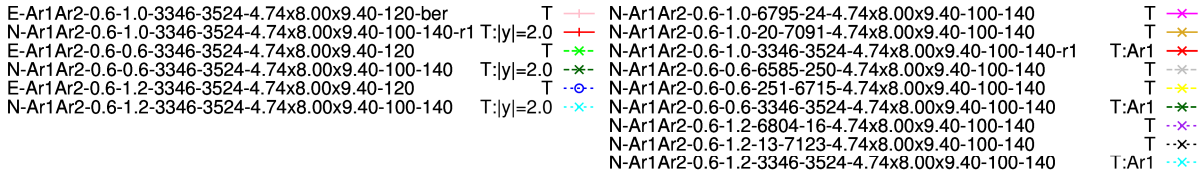

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:I
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 ρ:I
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 ρ:I
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1
N-Ar1Ar2-0.6-0.6-6585-250-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-0.6-251-6715-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.2-6804-16-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.2-13-7123-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140
∂(μ(ρ))/∂y [nm-4]
z [nm]
MeansLocDenYGradZ.eps
ø[μ(ρ)] [nm-3]
|y| [nm]
MeansLocDenMinY.eps

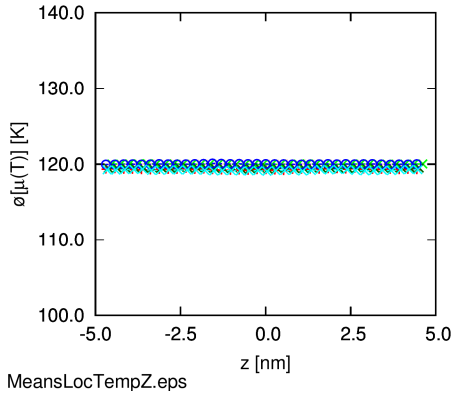

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:Ar1
N-Ar1Ar2-0.6-0.6-6585-250-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-0.6-251-6715-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
N-Ar1Ar2-0.6-1.2-6804-16-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.2-13-7123-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
ø[σ(ρ)] [nm-3]
z [nm]
SigmaLocDenZ.eps
ø[σ(ρ)] [nm-3]
|y| [nm]
SigmaLocDenOvMajY.eps



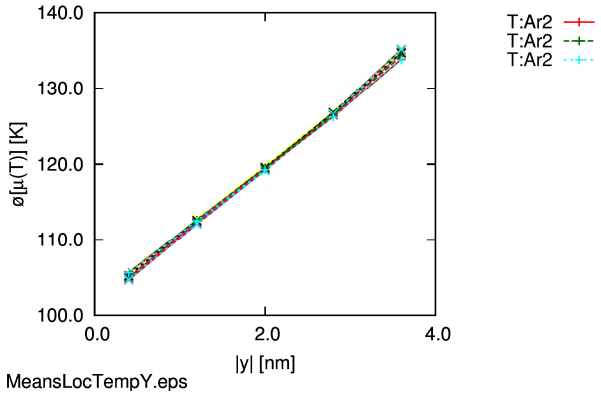
SigmaLocDenYGradZ.eps SigmaLocDenMinY.eps

a) Densities

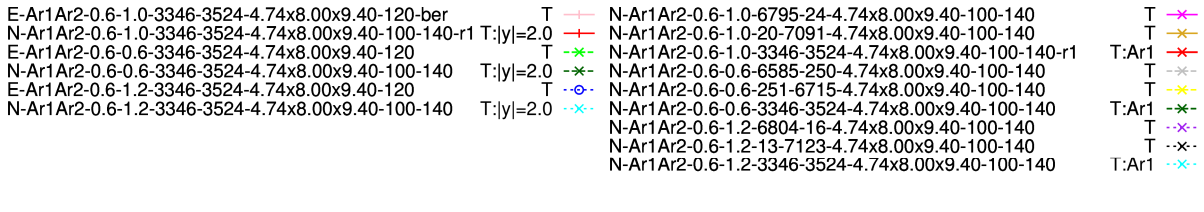


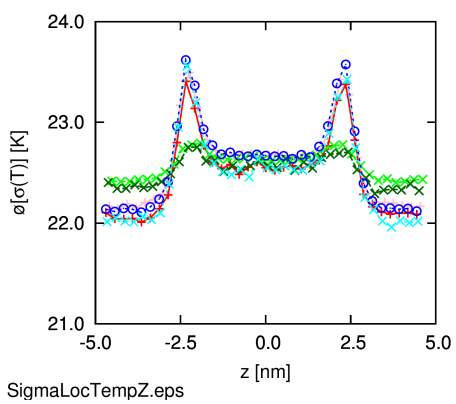


MeansLocTempZ.eps

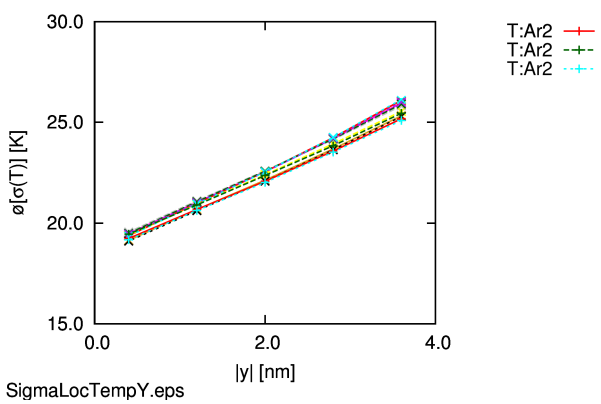


MeansLocTempY.eps

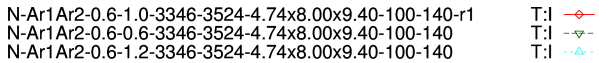


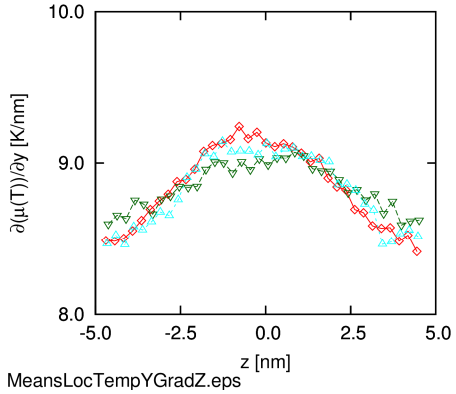


SigmaLocTempZ.eps



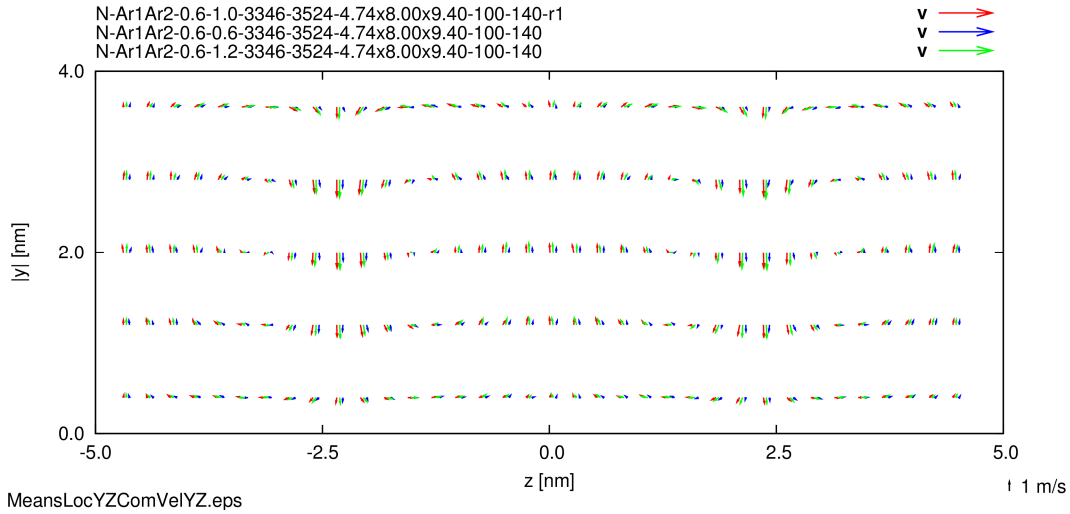
SigmaLocTempY.eps





MeansLocTempYGradZ.eps

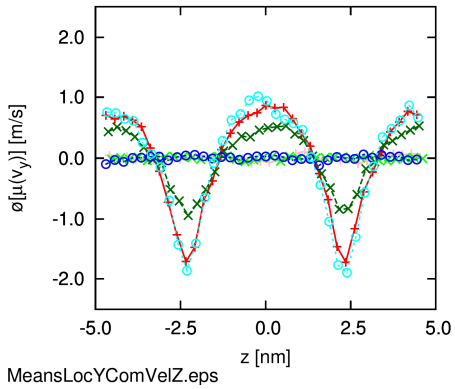
b) Temperatures



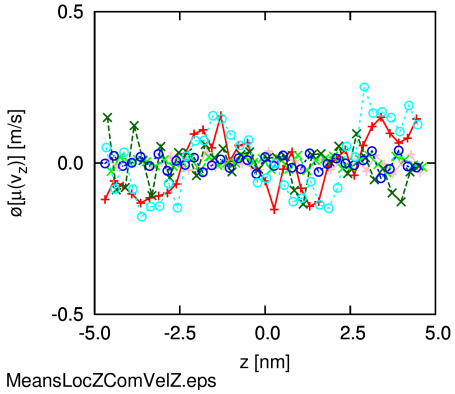
MeansLocYZComVelYZ.eps

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber v_y
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 v_y:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 v_y
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 v_y:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 v_y
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 v_y:|y|=2.0

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber v_z
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 v_z:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 v_z
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 v_z:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 v_z
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 v_z:|y|=2.0



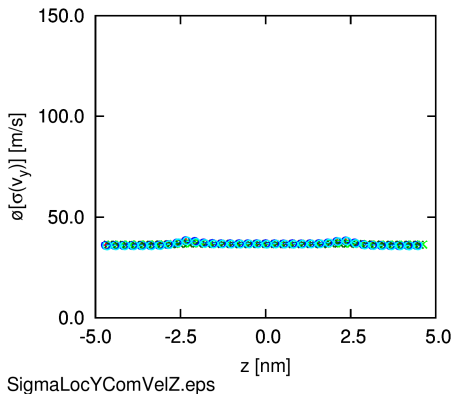
MeansLocYComVelZ.eps



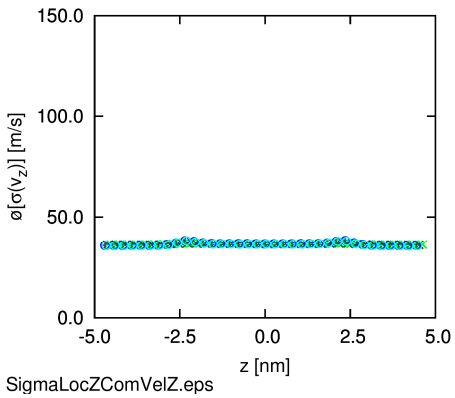
MeansLocZComVelZ.eps

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber v_y
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 v_y:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 v_y
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 v_y:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 v_y
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 v_y:|y|=2.0

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber v_z
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 v_z:|y|=2.0
E-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-120 v_z
N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140 v_z:|y|=2.0
E-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-120 v_z
N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 v_z:|y|=2.0

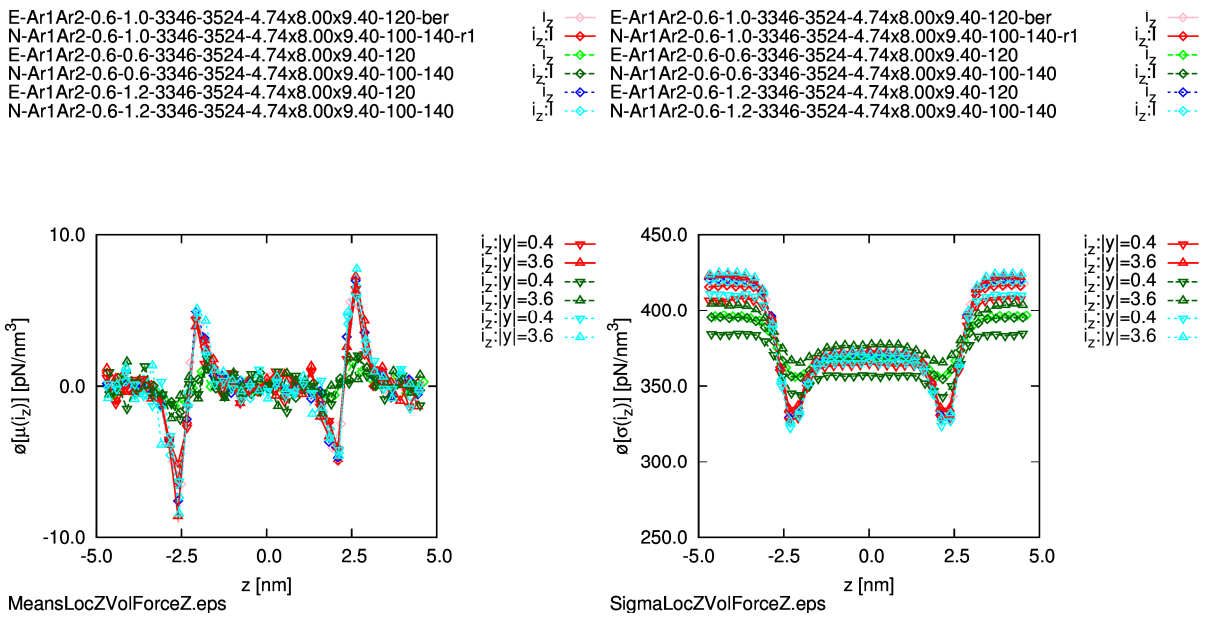


SigmaLocYComVelZ.eps



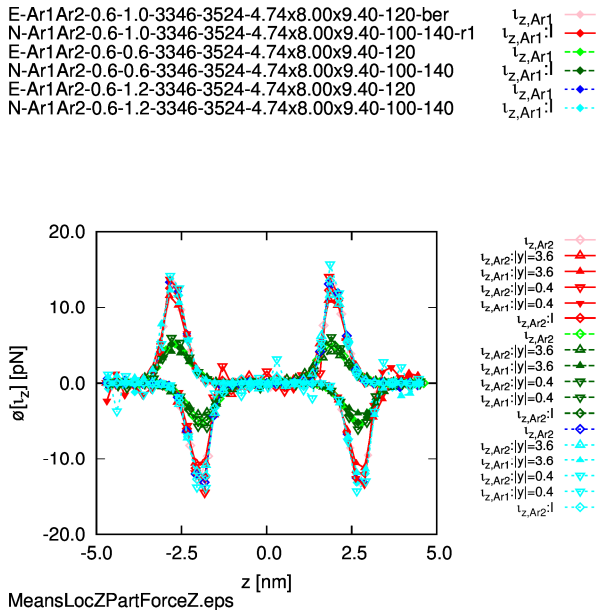
SigmaLocZComVelZ.eps

c) Com velocities



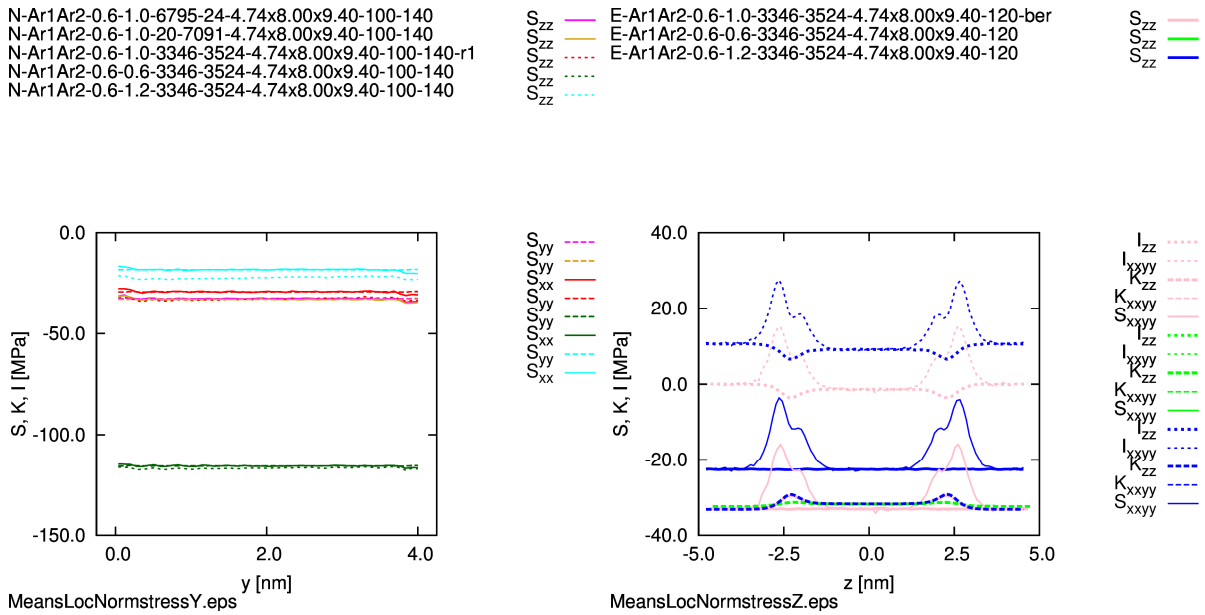
MeansLocZVolForceZ.eps SigmaLocZVolForceZ.eps

d) Volume forces



MeansLocZPartForceZ.eps

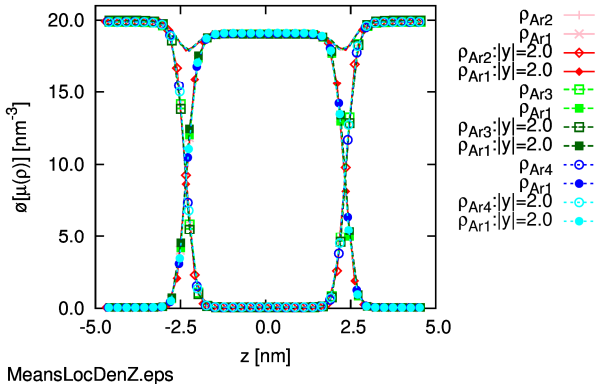
e) Particle forces



f) Normal stresses

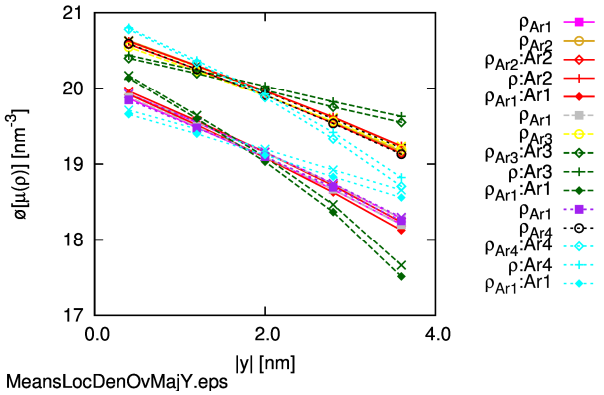
Figure 39 Comparison of the summarised local observables in several heterophasic nonequilibrium interfacial systems with different cut-off radii N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1, N-Ar1Ar2-0.6-0.6-3346-3524-4.74x8.00x9.40-100-140, and N-Ar1Ar2-0.6-1.2-3346-3524-4.74x8.00x9.40-100-140 We include also their corresponding systems in the comparison.

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:|y|=2.0
E-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
E-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0



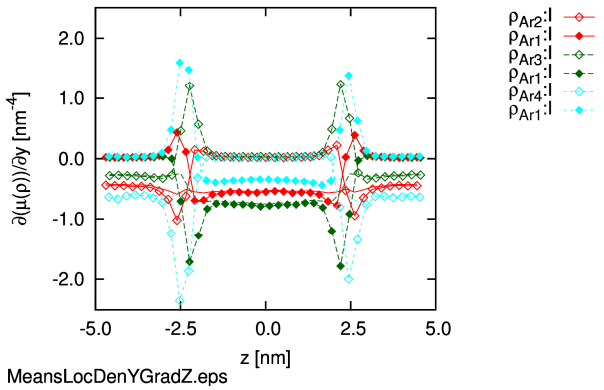
MeansLocDenZ.eps

N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:Ar1
N-Ar1Ar3-0.6-1.0-6797-22-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar3-0.6-1.0-20-7089-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
N-Ar1Ar4-0.6-1.0-6799-22-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar4-0.6-1.0-19-7088-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1



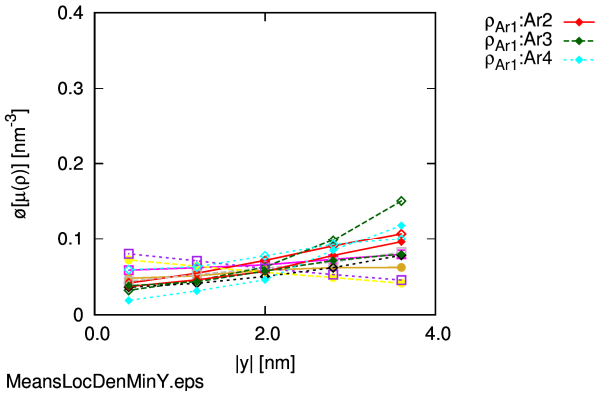
MeansLocDenOvMajY.eps

N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:I
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:I
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:I

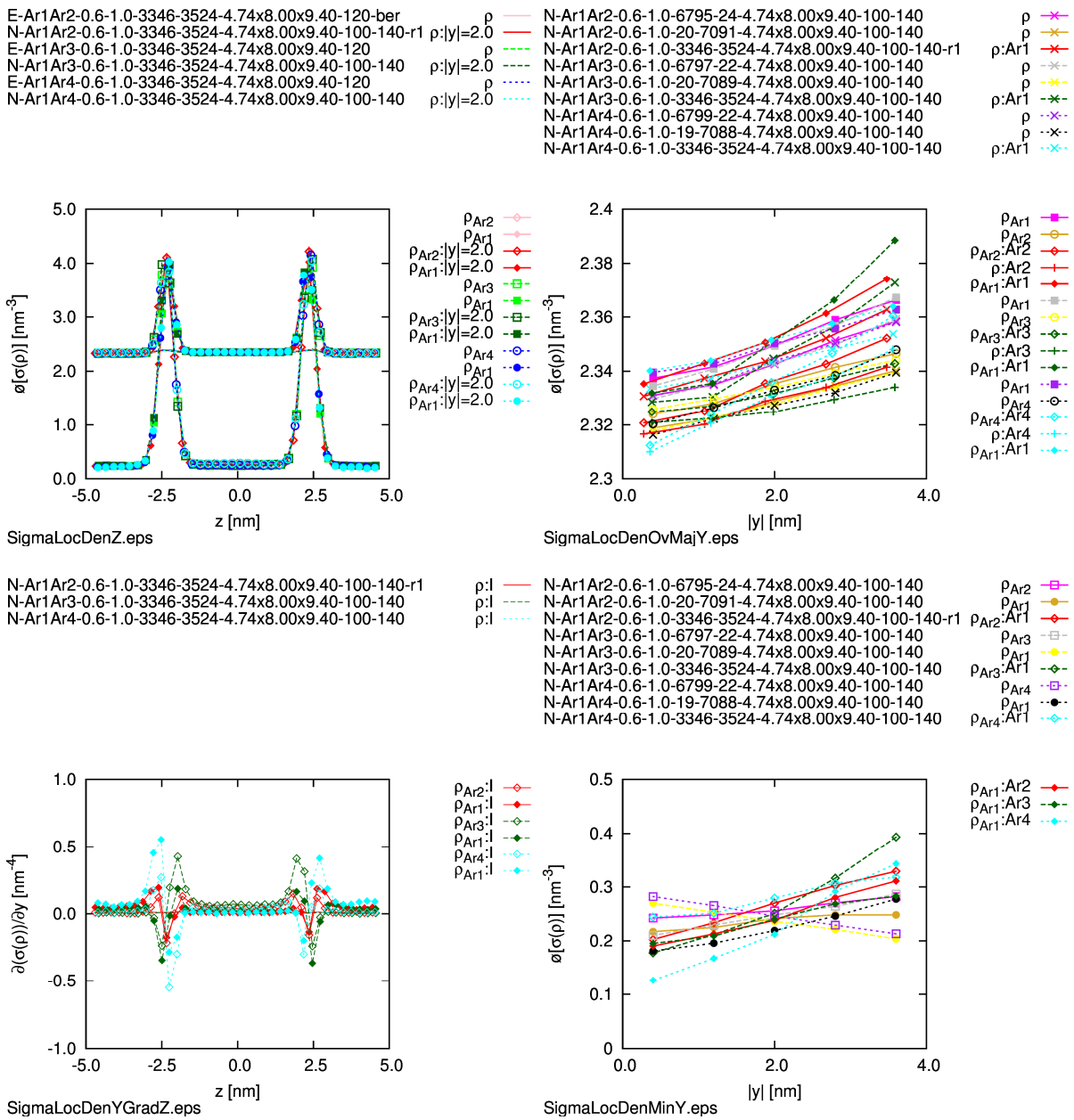


MeansLocDenYGradZ.eps

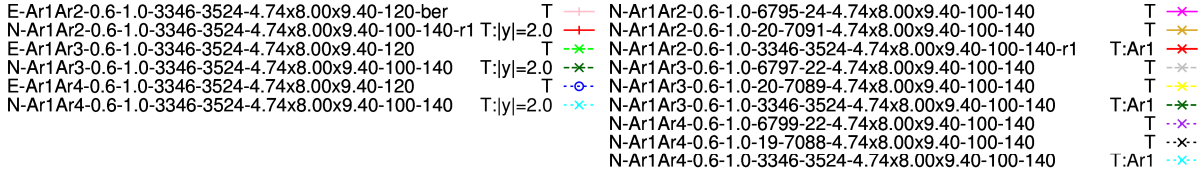
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ρAr2
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 ρAr1
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρAr2:Ar1
N-Ar1Ar3-0.6-1.0-6797-22-4.74x8.00x9.40-100-140 ρAr3
N-Ar1Ar3-0.6-1.0-20-7089-4.74x8.00x9.40-100-140 ρAr1
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρAr3:Ar1
N-Ar1Ar4-0.6-1.0-6799-22-4.74x8.00x9.40-100-140 ρAr4
N-Ar1Ar4-0.6-1.0-19-7088-4.74x8.00x9.40-100-140 ρAr1
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρAr4:Ar1

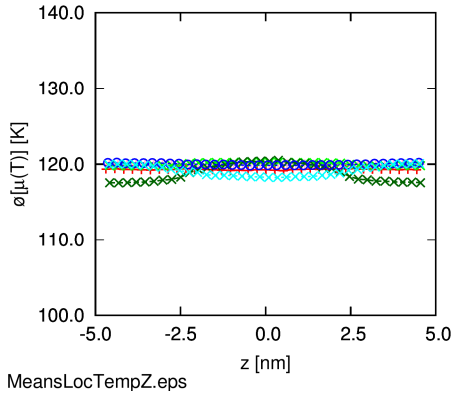


MeansLocDenMinY.eps

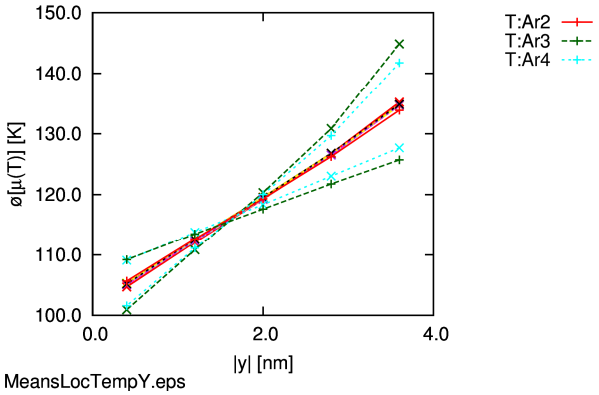

E-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-120-ber ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:|y|=2.0
E-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
E-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-120 ρ
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:|y|=2.0
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:Ar1
N-Ar1Ar3-0.6-1.0-6797-22-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar3-0.6-1.0-20-7089-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
N-Ar1Ar4-0.6-1.0-6799-22-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar4-0.6-1.0-19-7088-4.74x8.00x9.40-100-140 ρ
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:Ar1
ø[σ(ρ)] [nm-3]
z [nm]
ρAr2
ρAr1
ρAr2:|y|=2.0
ρAr1:|y|=2.0
ρAr3
ρAr1
ρAr3:|y|=2.0
ρAr1:|y|=2.0
ρAr4
ρAr1
ρAr4:|y|=2.0
ρAr1:|y|=2.0
SigmaLocDenZ.eps
ø[σ(ρ)] [nm-3]
|y| [nm]
ρAr1
ρAr2
ρAr2:Ar2
ρ:Ar2
ρAr1:Ar1
ρAr1
ρAr3
ρAr3:Ar3
ρ:Ar3
ρAr1:Ar1
ρAr1
ρAr4
ρAr4:Ar4
ρ:Ar4
ρAr1:Ar1
SigmaLocDenOvMajY.eps
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1 ρ:I
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:I
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140 ρ:I
N-Ar1Ar2-0.6-1.0-6795-24-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.0-20-7091-4.74x8.00x9.40-100-140
N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1
N-Ar1Ar3-0.6-1.0-6797-22-4.74x8.00x9.40-100-140
N-Ar1Ar3-0.6-1.0-20-7089-4.74x8.00x9.40-100-140
N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140
N-Ar1Ar4-0.6-1.0-6799-22-4.74x8.00x9.40-100-140
N-Ar1Ar4-0.6-1.0-19-7088-4.74x8.00x9.40-100-140
N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140
ρAr2
ρAr1
ρAr2:Ar1
ρAr3
ρAr1
ρAr3:Ar1
ρAr4
ρAr1
ρAr4:Ar1
∂(σ(ρ))/∂y [nm-4]
z [nm]
ρAr2:I
ρAr1:I
ρAr3:I
ρAr1:I
ρAr4:I
ρAr1:I
SigmaLocDenYGradZ.eps
ø[σ(ρ)] [nm-3]
|y| [nm]
ρAr1:Ar2
ρAr1:Ar3
ρAr1:Ar4
SigmaLocDenMinY.eps

a) Densities

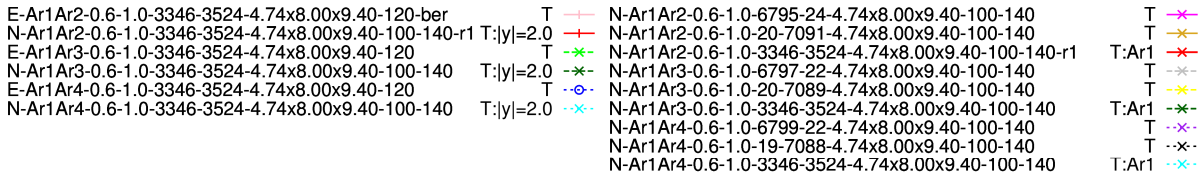


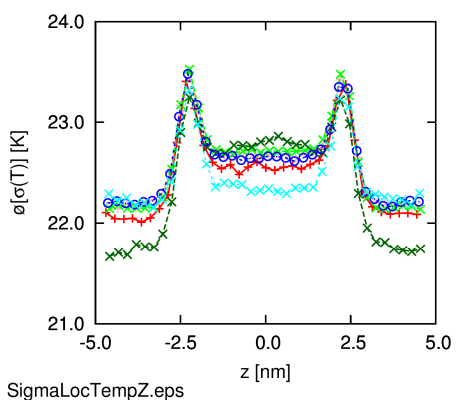


MeansLocTempZ.eps

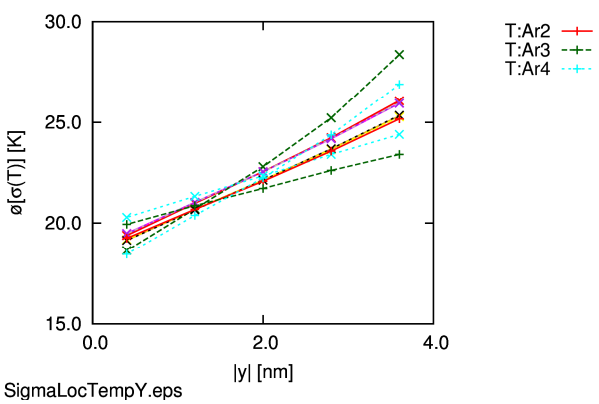


MeansLocTempY.eps

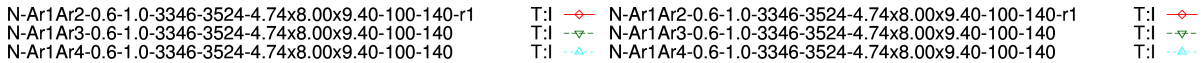


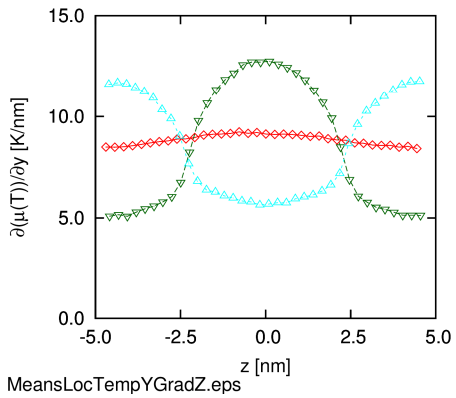


SigmaLocTempZ.eps

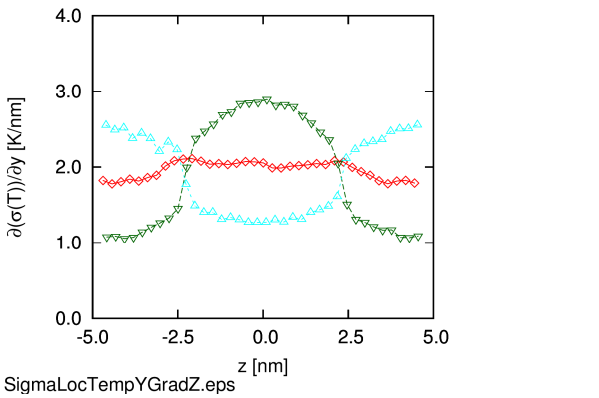


SigmaLocTempY.eps





MeansLocTempYGradZ.eps



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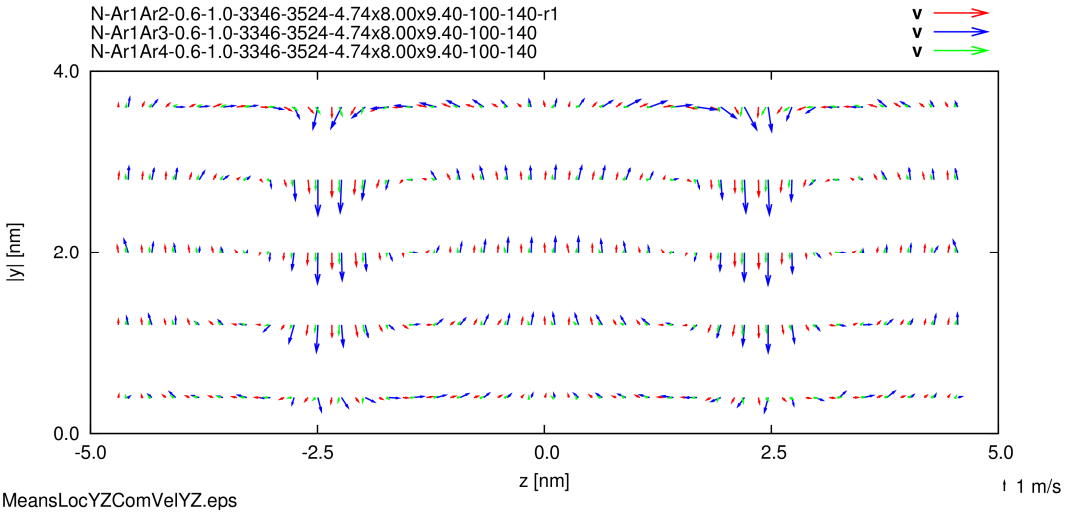
b) Temperatures



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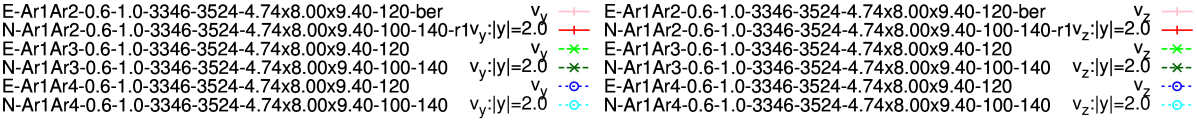

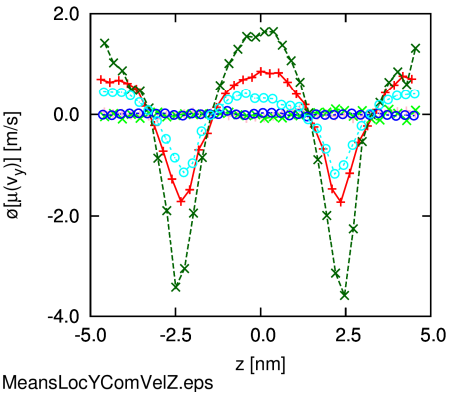

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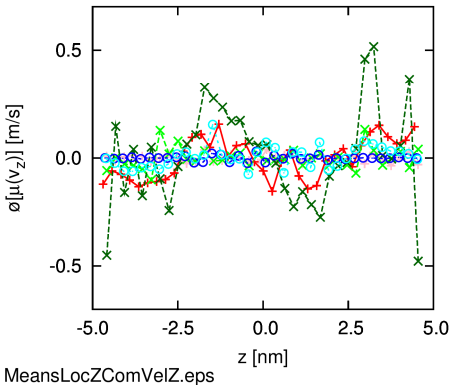

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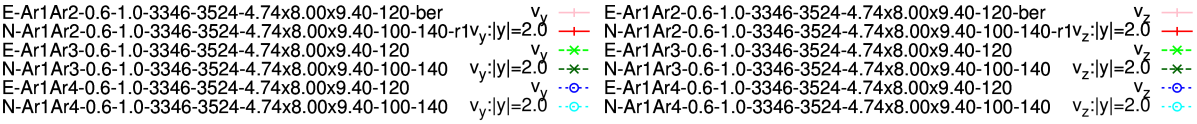

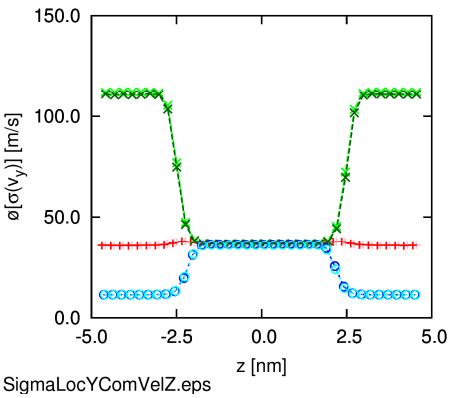

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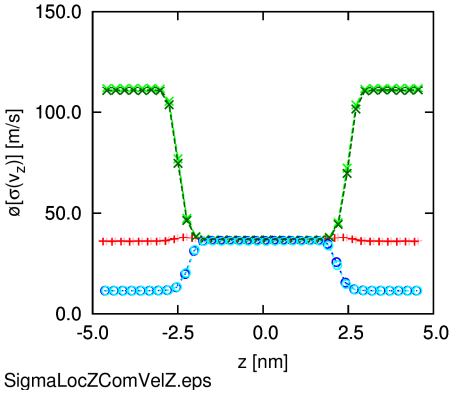
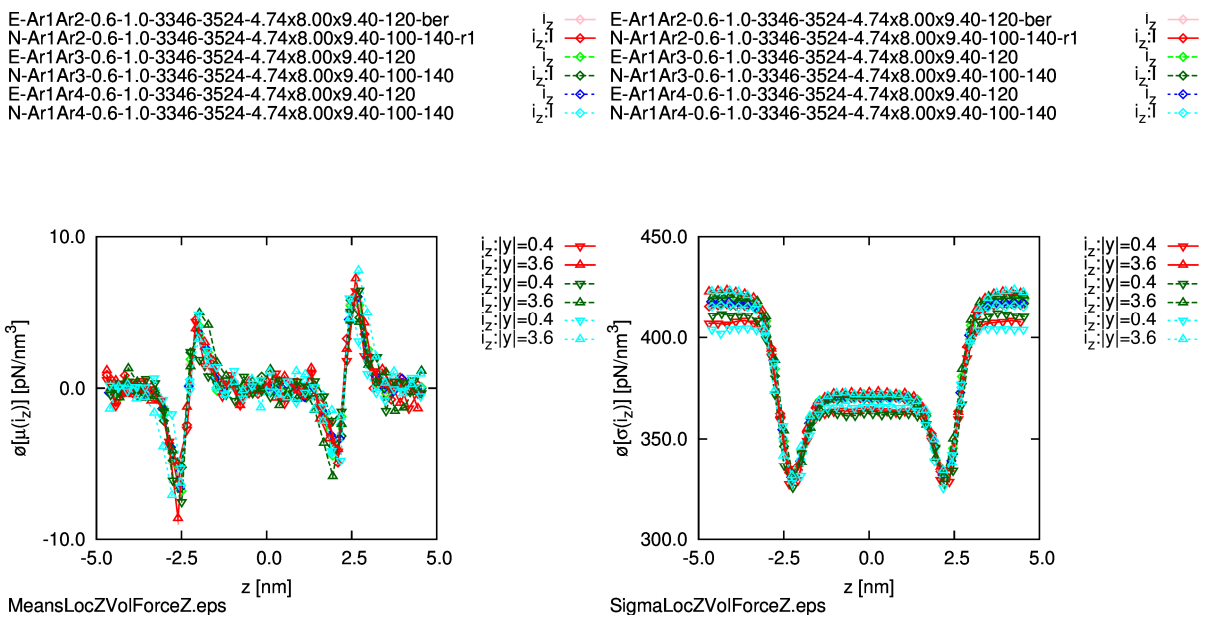
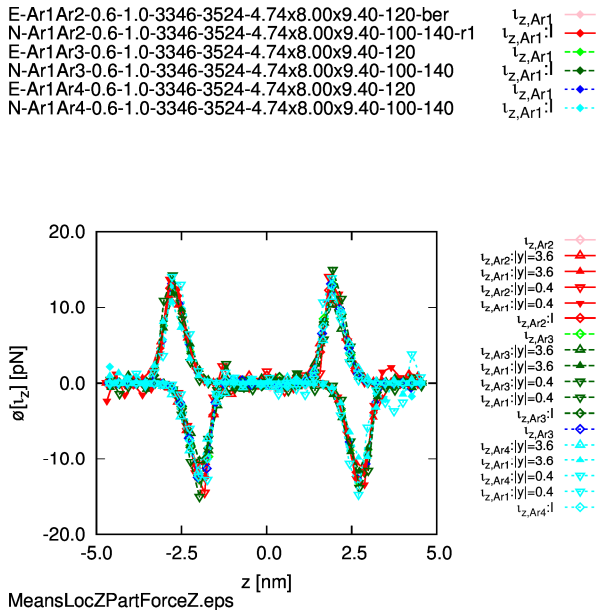


SigmaLocZComVelZ.eps

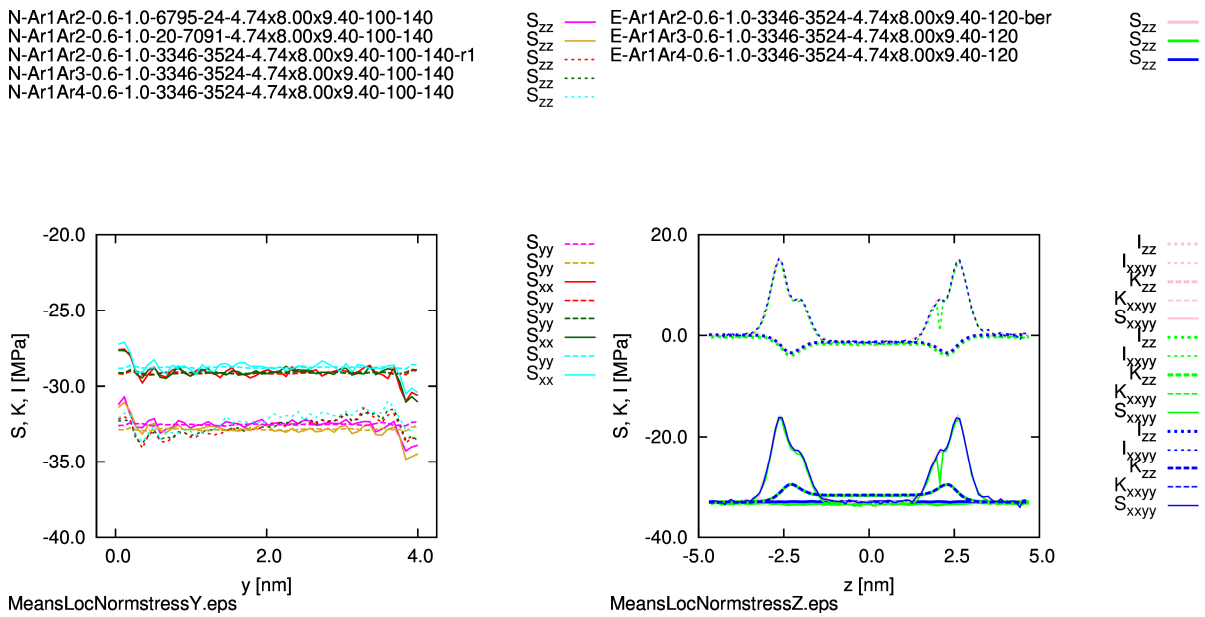
c) Com velocities



d) Volume forces

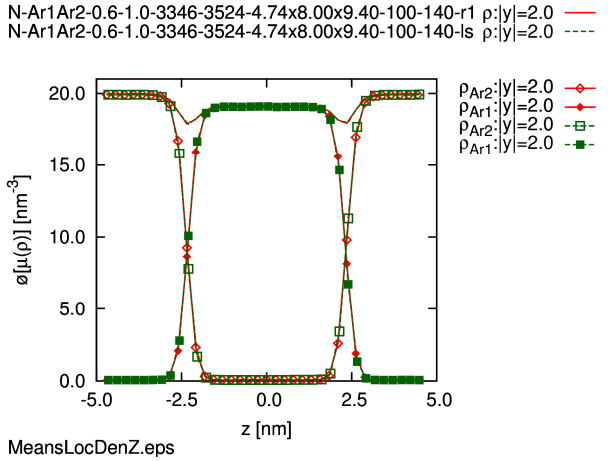


e) Particle forces



f) Normal stresses

Figure 40 Comparison of the summarised local observables in several heterophasic nonequilibrium interfacial systems with different ArB particle masses N-Ar1Ar2-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140-r1, N-Ar1Ar3-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140, and N-Ar1Ar4-0.6-1.0-3346-3524-4.74x8.00x9.40-100-140; We include also their corresponding systems in the comparison.



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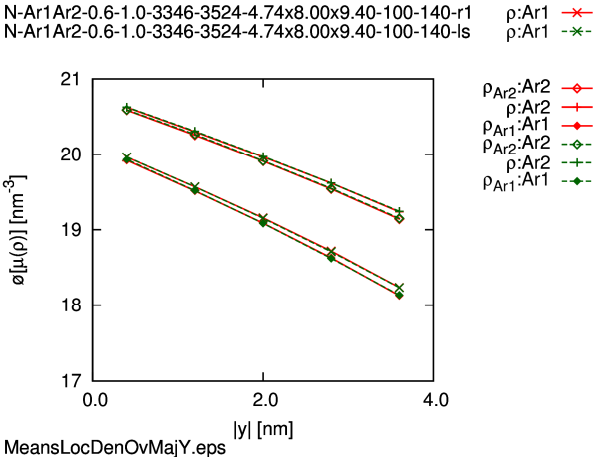

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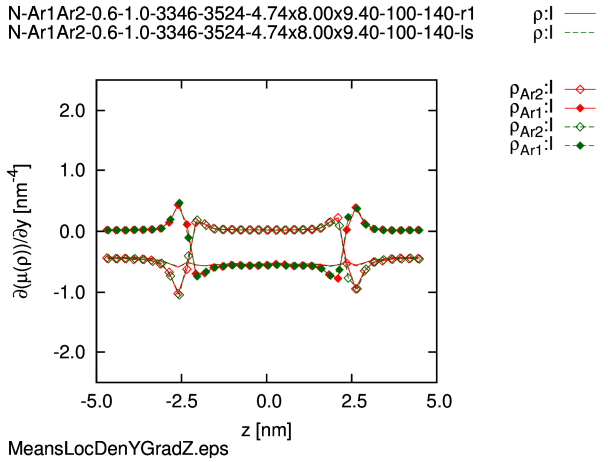
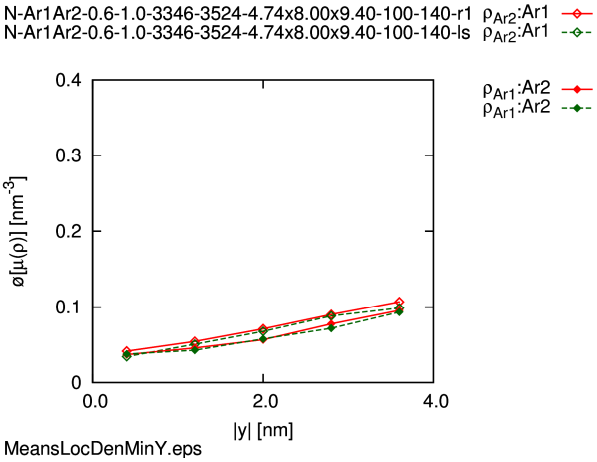


MeansLocDenYGradZ.eps



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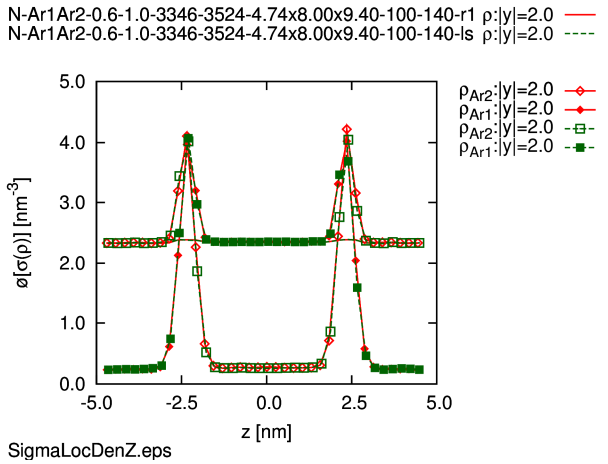

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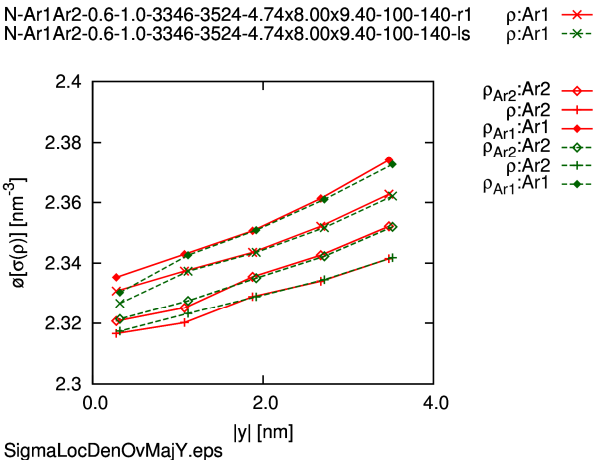

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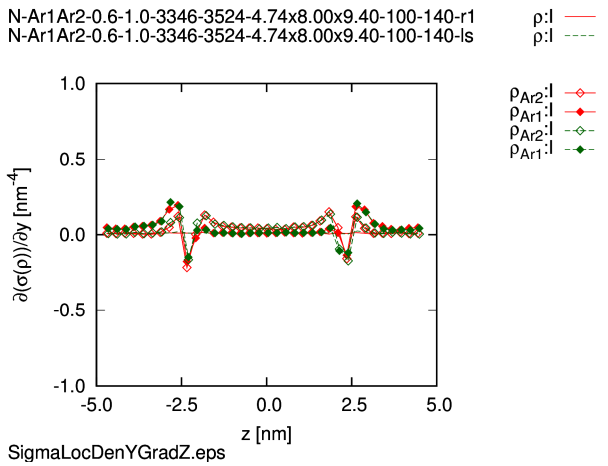

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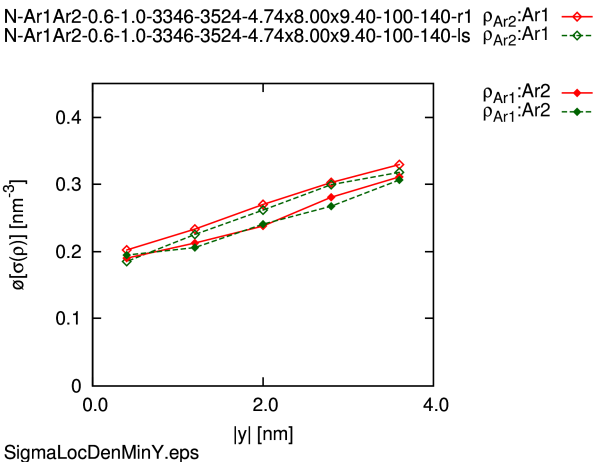

SigmaLocDenMinY.eps

a) Densities